July 26, 2017
Dear Editor,

We would like to thank you for serving as the editor for our manuscript. We have revised the manuscript to incorporate all of the general and specific comments and suggestions from the two reviewers. As the reviewers have suggested, we have removed section 3.5 and some sentences in other sections that appeared speculative, where we only had weak evidence to justify our arguments. Similarly, we rephrased several sentences and made them more understandable by adding further information. We believe that with these revisions the interpretation of the findings and hence the quality of the manuscript has improved significantly.

Sincerely yours,

Khadak Singh Mahata on behalf of all coauthors
We would like to thank both reviewers for their constructive comments and suggestions. Please find below the reviewers’ comments in black, our response in blue and changes in the revised manuscript in red. The line numbers in our response refer to the line numbers in the revised manuscript.

**Reviewer 1**

General comments: Atmospheric greenhouse gases (GHGs) such as CO2, CH4, H2O and CO are important climate forcing agents having significant impacts on climate system and air quality. This study brings outs first continuous measurements of atmospheric GHGs using high precision cavity ring down spectrometer (Picarro 24G2401, USA) at Kathmandu Valley during March 2013 to March 2014. The authors have done and extensive study on GHGs variability with time and space. However, there are a few minor technical changes in the manuscript. This paper is recommend to publish in ACP after incorporating the minor technical corrections.

We would like to thank reviewer for considering that the manuscript contains extensive work on the variability of GHGs in the Kathmandu Valley. We have incorporated the reviewer’s comments and suggestions to the extent possible in the following sections of the manuscript.

Line18-20: This paper studies about GHGs and GHGs are not classified as pollutants especially CO2 and CH4. Impact of pollution on GHGs need to be emphasis not to refer GHGs as pollutants. Also sentence “This paper reports …”. May be re-written

We agree with the reviewer that CO2 and CH4 at normal ambient concentrations are not considered as pollutants from the health perspective. Thus, we did not describe them as pollutants in the manuscript. We slightly modified the sentence “this paper reports…” to
distinguish the GHGs from the pollutant CO, and thus more understandably reflect what has been looked into in this study.

Line 51-53: Not clear. Authors may please check the sentence. “All three species showed strong diurnal and saying immediately CH4 and CO did not show any variation…”. May be provided quantitative numbers.

Thank you for pointing out that the statement was not clear to the reviewer. Here we are explaining the diurnal variations of CH4, CO2 and CO at two measurement sites (Bode and Chanban). We have articulated it by rephrasing the sentence as follows (lines 52-56):

At Bode, all three gas species (CO2, CH4 and CO) showed strong diurnal patterns in their mixing ratios with a pronounced morning peak (ca. 08:00), a dip in the afternoon, and again gradual increase through the night until the next morning, whereas CH4 and CO mixing ratios at Chanban did not show any noticeable diurnal variations.

Line 139: Rupakheti et al., 2016 need to be updated if available

The Rupakheti et al. manuscript will be submitted to ACPD soon. We have updated the citation as Rupakheti et al. (2017, manuscript in preparation) in line 144-145.

Line 252: Sentence “The % may be written as Difference (%) of the analyzer differed by…”. Corrected. The sentence has been rephrased as follows (lines 258-260):

The difference between CO2 mixing ratio reported by the analyzer and the reference mixing ratio was within 5%.

Line 349: Units of GHGs and other gases should be uniform in the manuscript.

Corrected. They are now reported in ppm throughout the manuscript.
Frequent rainfall suppresses emission sources which results in reducing mixing ratios of the gas and aerosol species. To reflect this, we have modified the sentences in lines 414-422:

The concentrations of most pollutants in the region are lower during the monsoon period (Sharma et al., 2012, Marinoni, 2013; Putero et al., 2015) because frequent and heavy rainfall suppresses emission sources. We saw a drop in the CO₂ mixing ratio during the rainfall period due to changes in various processes such as enhanced vertical mixing, uptake of CO₂ by vegetation and soils, and where relevant reduction in combustion sources. CO₂ can also dissolve into rainfall, forming carbonic acid, which may lead to a small decrease in the CO₂ mixing ratio as has been observed during heavy intensity rainfall (Mahesh et al., 2014; Chaudhari et al., 2007).

The following references are added in the list of references.


Line 432-433: Please check the statement that CO₂ will be high but CH₄ will be high during post-monsoon season.

This comment does not match with the lines mentioned by the reviewer. We did not find the exact or similar sentence in the manuscript.
Figure 4: Titled should be changed. Since GHGs are not pollutants and legend should be CH4 not Ch4.

In order to remove confusion, we have changed the caption of Figure 4 as follows:

Relation between mixing ratios and wind direction observed at Bode in the Kathmandu Valley (a) CH4, (b) CO2, and (c) CO from March 2013 to February 2014. The figure shows variations of CH4, CO2 and CO mixing ratios based on frequency counts of wind direction (in %) as represented by circle. The color represents the different mixing ratios of the gaseous species. The units of CH4, CO2 and CO are in ppm.

The legend is also corrected as suggested.

Figure 11: Show double Y-axis for better visualization

Thank you for the suggestion. As individual species varies over a wide range and there is also an order of magnitude difference in their mixing ratios, putting them in double Y-axes with different scales makes the figure more confusing. We have therefore kept the figure as it is.

Reviewer 2

Highly precise and long-term measurements of greenhouse gases are essential to understand the underlying processes in the context of global climate change. It is particularly valuable in regions like Asia where it is currently limited by dedicated long term observations. This study demonstrates the observational variations of CO2 and CH4 mixing ratios in the Kathmandu Valley (Nepal), and compares them to those of other rural sites. The scope of this study is hence highly relevant to the public and authors’ efforts on this regard need to be appreciated. However, the manuscript in its present form does not meet the standard to merit the publication in ACP, and needs to be adequately revised. I therefore recommend the current manuscript to undergo major revision to be considered in ACP.
We would like to thank reviewer for considering our measurements as important for the region and that the scope of our study is relevant to the public. We have revised the manuscript as suggested by the reviewer.

General comments
As mentioned above, I value authors’ effort in this study as an important step towards generating observational wealth which can be tremendously used by scientific community to understand a wide range of mechanisms involved in this aspect. Given the complicated interplay of many processes involved, single approach cannot answer the unresolved scientific questions related to these processes and mechanisms affecting mixing ratio variations. This requires defining far more systematic approaches and other robust tools. However the finding from this study can be very valuable if presented with adequate measurement and analysis methods/techniques used, including a good summary of the methods and a much clear report on the observational variations of these analysed tracers. The results will be more convincing by focusing on the key aspects of the data rather than trying to relate them to flux categories based on assumption (many times) and without sufficient tools (inverse modeling). In some places, results are presented nicely, but then an explanation is suggested based on other literature that focussed on other study regions/methods, without even demonstrating its relevance to this dataset. Also in some places, the study gets into overly ambitious interpretations of the results and conclusions on the basis of single analysis. On this basis, I recommend authors to focus on presenting this study by clearly stating the measurement and analysis techniques used, defining their strategies, providing analysed results & possible uncertainties and giving more convincing interpretations of the results and conclusions. I highly recommend authors not to jump to interpreting emission/flux sources and patterns based on the single site measurements and the analysis done in this study. Sections in this manuscript dealing with these aspects needs to be (preferably) removed or highly restructured.

Thank you for the general comments. We would like to break down the general comments, as suggested by the editor, to address reviewer’s general comments systematically and clearly as follows:
As mentioned above, I value authors’ effort in this study as an important step towards generating observational wealth which can be tremendously used by scientific community to understand a wide range of mechanisms involved in this aspect.

Thanks for considering our work as observational wealth that can be useful for the scientific community to understand the process involved in this aspect.

Given the complicated interplay of many processes involved, single approach cannot answer the unresolved scientific questions related to these processes and mechanisms affecting mixing ratio variations. This requires defining far more systematic approaches and other robust tools. However the finding from this study can be very valuable if presented with adequate measurement and analysis methods/techniques used, including a good summary of the methods and a much clear report on the observational variations of these analysed tracers. The results will be more convincing by focusing on the key aspects of the data rather than trying to relate them to flux categories based on assumption (many times) and without sufficient tools (inverse modeling).

We agree with this comment that a single approach in our study cannot answer the resolved scientific questions. We apologize for not mentioning the approaches clearly. We have improved measurement and analysis methods as: (i) resolved the confusion between uncertainty and variability (expressed as standard deviation) and recalculated annual values of all species from monthly data to make them comparable with background and regional sites, as explained in the specific comments below (also refer to lines 316-322 in the manuscript), (ii) performed significance test of CH4 and CO2 data between Bode and Mauna Loa, and Bode and Waliguan. Similarly, in the results part, we have removed (i) the loose connection to regional pollution in section 3.5 (influence of regional emission and transport) and (ii) substantially revised section 3.6 (CO and CO2 ratio: potential emission sources), clearly mentioning that the information on sources inferred from the ratio analysis is only indicative rather than definitive (lines 680-682 and lines 715-730) and (iii) also included useful statistics in Table 6.
In some places, results are presented nicely, but then an explanation is suggested based on other literature that focussed on other study regions/methods, without even demonstrating its relevance to this dataset.

We have selected other urban/semi-urban sites in India as mentioned in the manuscript on the basis of similarity of emission sources and similar influence of regional meteorology in the region (as mentioned in lines 325-328). The background sites (Mauna Loa and Waliguan) were selected to see the spatial difference between the background sites and current study sites as described in lines 322-325 in the manuscript.

Also in some places, the study gets into overly ambitious interpretations of the results and conclusions on the basis of single analysis.

We would like to thank reviewer for noticing these ambitious interpretations. We have rephrased such interpretations, mainly in the lines 715-730 and lines 835-837.

On this basis, I recommend authors to focus on presenting this study by clearly stating the measurement and analysis techniques used, defining their strategies, providing analysed results & possible uncertainties and giving more convincing interpretations of the results and conclusions. I highly recommend authors not to jump to interpreting emission/flux sources and patterns based on the single site measurements and the analysis done in this study. Sections in this manuscript dealing with these aspects need to be (preferably) removed or highly restructured.

While revising the manuscript, We have focused on the key aspect of the data: (i) better interpretation of the results, as suggested by the reviewer, and (ii) removed or refined speculative sentences and (iii) removed the whole section 3.5 (influence of regional emission and transport), which the reviewer found less convincing. Also we have substantially revised the interpretation of emission sources based on CO/CO₂ ratios, clearly stating that the results are only indicative, not confirmative (lines 715-730). In addition to the points noted above, we hope that by
addressing all the specific comments collectively we have adequately addressed the referee’s general comments as well.

Specific comments and suggestions for revised analysis
Page 3, lines 63-66 “Between 1750…..cover changes” Misleading sentence. The given estimation is overly high for the accumulated CO2 in the atmosphere. As per IPCC reports & other studies, it is in the range of 230-250 Pg C. The given number is more towards total (cumulative) CO2 emissions between 1750 and 2011, which were partly compensated by the ocean and terrestrial ecosystems.

We would like to thank reviewer for noticing the misleading sentence which is now corrected with CO2 accumulation data from the IPCC 2013 report as follows (lines 66-67):

Between 1750 and 2011, 240(±10) PgC of anthropogenic CO2 was accumulated in the atmosphere…. (IPCC, 2013).

Page 4, line 86-89 “important sources” Give reference

The following papers have been cited (see changed line 90-94) and the full citation is also included in the reference section.


Page 4, lines 90-92 “Ecosystem and.... between July-October Please give appropriate reference for this statement. Prasad et al., 2014 investigated based on satellite GHG concentration observations, not based on inverse or ecosystem models. In my knowledge there’s no such inverse flux estimations available over South Asia, accounting (and decoupling) seasonal variations of CO2 uptake and release to the atmosphere. However, Patra et al., 2011 showed inverse estimations of monthly co2 fluxes over South Asia. Please correct it.

Thank you for pointing out the incorrect statement. The sentences and reference have been replaced by relevant reference in lines 95-100 as follows:

By using inverse modeling, Patra et al. (2011) found a net CO₂ uptake (0.37 ± 0.20 Pg C yr⁻¹) during 2008 in South Asia and the uptake (sink) is highest during July-September. The remaining months act as a weak gross sink but a moderate gross source for CO₂ in the region.

Page 12, lines 308-310 “2.193 (_ 0.224) ppm, 419.4 (_ 23.9) ppm, 0.50 (_ 0.35) ppm, and 310 1.71 (_ 0.71) %” Uncertainty seems to be quite high for co2, ch4 and co. Why? Please include the reason in the text.

We apologize for not clarifying about the values in the parenthesis adjacent to the annual average value for the three species. They are not measurement uncertainties. Instead, they are one standard deviation, calculated from the hourly data for the observation period (of one year). The reported annual mean from the references sites such as Mauna Loa and referred to in this paper were calculated from monthly means, not hourly data. Therefore for consistency and ease of comparison, we have now reported annual average and the standard deviation from monthly mean data for all sites. Please see the correction (lines 316-322):

For the entire sampling period, the annual average (± one standard deviation) of CH₄, CO₂, CO, and water vapor mixing ratios at Bode were 2.192 (± 0.066) ppm, 419.3 (± 6.0) ppm, 0.50 (± 0.23) ppm, and 1.73 (± 0.66)% respectively. The relative variabilities for CH₄, CO₂ and CO were thus 3%, 1.4% and 46%, respectively. Their variabilities at Mauna Loa were CH₄: 6% and CO₂: 0.5% and at Waligaun were CH₄: 0.48%, CO₂: 0.9%. The high variability in the annual mean,
notably for CO in Bode could be indicative of the seasonality of emission sources and meteorology.

Page 12, lines 318-320 “CH4 was ….. observation period” Given the above uncertainty ranges of Bode values (nearly 10.2% for CH4, 5.7% for CO2 and 70% for CO), the estimated percentages of increment relative to other observatories are also biased. These uncertainties need to be taken into account, or at least properly mentioned.

As noted above, these are not uncertainties, rather variabilities. We have recalculated the annual average and standard deviation values based on monthly average data. Now, the variabilities for CH4, CO2 and CO at Bode are 3%, 1.4% and 46%, respectively. The comparison between Bode and the other sites (Mauna Loa and Waligauan) for the GHGs now looks more reasonable. The following text has been inserted to reflect their variabilities and statistical significance (lines 330-334)

We performed a significance test at 95% confidence level (t-test) of the annual mean values between the sites to evaluate whether the observed difference is statistically significant (p < 0.05), which was confirmed for the annual mean CH4 and CO2 between Bode and Mauna Loa, and between Bode and Waliguan.

Page 12, lines 320-322 “The small .... Asia region” Although I tend to agree that we can expect (+ seeing satellite images), most of the cases, higher CH4 mixing ratios in Asia relative to Mauna Loa observatory, authors should note that this conclusion, as given in the text, about the whole Asia cannot be drawn from analysing “just two Asian sites” in a given time period. This sentence is misleading, and needs to be reformulated.

Thanks for drawing our attention to it. Yes, we didn’t intend to generalize our comparison for Asia. We rephrased the following paragraph (lines 334-340):

CH4 was nearly 20% higher at Bode than at Mauna Loa (1.831 ± 0.110 ppm) (Dlugokencky et al., 2017) and ca.17% higher than at Mt. Waliguan (1.879 ± 0.009 ppm) for the same observation
period (Dlugokencky et al., 2016). The slightly higher CH$_4$ mixing ratios at Bode and Waliguan than at Mauna Loa Observatory could be due to prevalence of rice farming as a key source of CH$_4$ in this part of Asia.

Page 12, line 324 “Ahmedabad (1.880 ppm) (Sahu and Lal, 2006) and Shadnagar (1.92 ± 0.07)” See my above comment on uncertainty.

We have calculated the variabilities for Bode, Ahmedabad and Shadnagar, and rephrased the sentences in lines 340-345 as follows:

Similarly, the annual average CH$_4$ at Bode during 2013-14 was found comparable to an urban site in Ahmedabad (1.880 ± 0.4 ppm, i.e., variability: 21.3%) in India for 2002 (Sahu and Lal, 2006) and 14% higher than in Shadnagar (1.92 ± 0.07 ppm, i.e., variability: 3.6%), a semi-urban site in Telangana state (~70 km north from Hyderabad city) during 2014 (Sreenivas et al., 2016).

Page 12, lines 326-332 “Likewise, the .... in China” These estimated increments are meaningless based on the uncertainty range of Bode’s tracer mixing ratios. That is, these 5.7% and 5.5% increments are statistically insignificant when it is compared with CO2 values with 5.7% uncertainty range. I strongly recommend authors to remove this.

See above regarding the confusion between uncertainty and variability, for which we apologize that this was not clear. We have recalculated the standard deviations based on the monthly values, and have also conducted a t-test between the annual mean mixing ratios and found that differences between the means at Bode and Mauna Loa, and between Bode and Waliguan were statistically significant. We have removed unnecessary explanations and inserted the following sentence (lines 345-348):

Likewise, the difference between annual mean CO$_2$ mixing ratios at Bode (419.2 ±6.0 ppm, 1.4% variability) vs. Mauna Loa (396.8 ± 2.0 ppm, 0.5% variability) (NOAA, 2015) and Bode vs. Waliguan (397.7 ± 3.6 ppm, 0.9% variability) (Dlugokencky et al., 2016a) is statistically significant (p <0.05).
Similar to Chanban, the burning activities around Bode area were also reduced or absent during the rainy season. We added the following line (378-379) in the manuscript.

The garbage and agro-residue burning activities were also absent or reduced around Bode due to rainfall during the monsoon period.

Agree. The paragraph has been restructured in lines 408-414 as:

The seasonal variation in CO$_2$ could be due to (i) the seasonality of major emission sources such as brick kilns, (ii) seasonal growth of vegetation (CO$_2$ sink) (Patra et al., 2011) and (iii) atmospheric transport associated with regional synoptic atmospheric circulation (monsoon circulation and westerly disturbance in spring season) which could transport regional emission sources from vegetation fires and agriculture residue burning (Putero et al., 2015), and a local mountain-valley circulation effect (Kitada and Regmi, 2003; Panday et al., 2009).

Page 14, lines 388 “partially due to rain washout. “ It can also very well due to relatively high advection and vertical mixing.

Thank you for pointing out other possible reasons of low mixing ratios of gas species in rainy season. They are included (lines 414-422):

The concentrations of most pollutants in the region are lower during the monsoon period (Sharma et al., 2012, Marinoni, 2013; Putero et al., 2015) because frequent and heavy rainfall
suppresses emission sources. We saw a drop in the CO₂ mixing ratio during the rainfall period due to changes in various processes such as enhanced vertical mixing, uptake of CO₂ by vegetation and soils, and where relevant reduction in combustion sources. CO₂ can also dissolve into rainfall, forming carbonic acid, which may lead to a small decrease in the CO₂ mixing ratio as has been observed during heavy intensity rainfall (Mahesh et al., 2014; Chaudhari et al., 2007).

Page 15, lines 395-396 “related to less or no rainfall, which results in the absence of rain washout” if it is with transport or raining, it should also affect CH₄ and other tracers. Please clarify.

The additional source of CH₄ is due to agricultural activities from the paddy fields in the monsoon season (esp. August-September) which is absent from October. Therefore we see the drop in CH₄ starting October. It is likely that the absence of rainfall after October is conducive to CO₂ accumulation and thus we see an increase in the mixing ratio of CO₂ thereafter. We have tried to clarify it in lines 432-433 as:

However, the reduction in ambient CH₄ after October could be due to reduced CH₄ emissions from paddy fields, which were high in August-September.

Page 15, lines 412-413 “which had > 2.5 ppm CH4 and > 450 CO2” It’s likely that the increase in CO2 & CH4 is associated with advected signals from the North East and the East; however it is not clear how it can be interpreted as the advected airmass had the said values for CO2 and CH4 unless the study used any tracer transport and emission models. Wind direction and tracer concentration from the given site alone are not sufficient to conclude this. A clarification is needed here. Otherwise I recommend authors to remove this. Page 15, lines 414 “(not shown in Figure 4)” I encourage authors to show CO as well for the completeness of the interpretation.

We agree that in the absence of tracer transport and emission modeling, it is not sufficient to conclude the directionality of the advected signals. The sentence formulation also gave an
impression that we are definitive about our conclusion. We have revised the lines 448-452. And CO has been included in Figure 4 as suggested by reviewer.

In the absence of tracer model simulations, the directionality of the advected air masses is unclear. Figure 4 shows that during these two months, CO₂ mixing ratios were particularly high (> 450 CO₂ and > 2.5 ppm CH₄) with the air masses coming from the Northeast-East (NE-E).

Page 15, lines 417 “high CH₄ emissions” What about CO₂?
The CO₂ level is lower during monsoon period in Asia due to high uptake of CO₂ by plants (high photosynthetic activities). This has been mentioned in lines 95-100 and 422-423 in the manuscript with reference.

Page 17, line 455 “The westerly circulation (originated at longitude about 60E in 5 days back trajectories)” What does it mean? What’s originated? Talking about model? Based on modeled trajectories?

The air mass back trajectory analysis is based on the study by Putero et al., 2015 for a site in the Kathmandu Valley for the period February 2013-January 2014. Using the Hysplit trajectory model, Putero et al (2015) clustered/grouped the 5 day back trajectories into a total of 9 clusters. The “westerly circulation” referred to in the manuscript is one of the dominant clusters (21.4%) observed by Putero. If spatially viewed on a lambert conformal projection, the majority of individual trajectories in the westerly circulation “or westerly cluster” originates in the region of 20-40°N, ~60°E.

We acknowledge that the sentence was confusing. Please see the following correction which, we hope, provides a fairly detailed and clear explanation (lines 495-504).

To relate the influence of synoptic circulation with the observed variability in BC and O₃ in the Kathmandu Valley, 5-day back trajectories (of air masses arriving in the Kathmandu Valley) were computed by Putero et al., (2015) using the HYSPLIT model. These individual trajectories which were initialized at 600 hPa, for the study period of one year, and were clustered into nine
clusters. Of the identified clusters, the most frequently observed clusters during the study period were the Regional and Westerly cluster or circulation (22% and 21%). The trajectories in the regional cluster originate within 10° x 10° around the Kathmandu Valley, whereas the majority of trajectories in this westerly cluster originated broadly around 20-40° N, ~60° E. Putero et al (2015) found that the regional and westerly synoptic circulation were favorable for high values of BC and O₃ in the Kathmandu Valley.

Page 17, lines 475-476 “CO₂ mixing ratios whereas CO shows an evening peak” It’s surprising. Why is it so? Please clarify.

We meant to say that the CO₂ and CH₄ keep increasing over evening time until early morning while CO shows a peak coinciding with evening peak traffic hour and then drops. The decay in CO is more pronounced in monsoon and post-monsoon seasons. We have tried to clarify further in lines 526-534.

The gradual increase of CO₂ and CH₄ mixing ratios in the evening in contrast to the increase until evening peak traffic hours and later decay of CO may be indicative of a few factors. As pointed out earlier, after the peak traffic hours, there are no particularly strong sources of CO, especially in the monsoon and post-monsoon season. It is also likely that some of the CO decay is due to nighttime katabatic winds which replace polluted air masses with cold and fresh air from the nearby mountain (Panday and Prinn, 2009). As for the CO₂, the biosphere respiration at night in the absence of photosynthesis can add additional CO₂ to the atmosphere, which especially in the very shallow nocturnal boundary layer may explains part of the increase of the CO₂ mixing ratio.

Page 19, lines 519-521 “While the .... other seasons” I couldn’t follow how it’s related. A clarification is highly needed. What are these other most CO sources mentioned here?

We have tried to make it clear and also included other CO sources in lines 578-581. The new sentences read as:
While the biosphere respires at night, which may cause a notable increase in CO₂ in the shallow boundary layer, most CO sources (transport sector, residential cooking) except brick kilns remain shut down or less active at night.

Page 20, lines 551-559 “Highest day ....Mauna Loa and Waliguan” It’s lost. I see many assumptions here rather than convincing statements. What about biospheric activity and its seasonality? mesoscale transport mechanisms?

We acknowledge that the whole paragraph was not clear. We have tried to make it concise and clear and also included some of your suggestions. The paragraph has been revised as follows (lines 612-628):

The highest daytime minimum of CO₂ was observed in the pre-monsoon, followed by winter (Figure 6b). The higher daytime minimum of CO₂ mixing ratios in the pre-monsoon season than in other seasons, especially winter, is interesting. The local emission sources are similar in pre-monsoon and winter and the boundary layer is higher (in the afternoon) during the pre-monsoon (~1200 meters) than in winter (~900 meters) (Mues et al., 2017). Also, the biospheric activity in the region is reported to be higher in the pre-monsoon (due to high temperature and solar radiation) than winter (Rodda et al., 2016). Among various possible causes, transport of CO₂ rich air from outside the Kathmandu Valley has been hypothesized as a main contributing factor, due to regional vegetation fire combined with westerly mesoscale to synoptic transport (Putero et al. 2015). In monsoon and post-monsoon seasons, the minimum CO₂ mixing ratio in the afternoon drops down to 390 ppm, this was close to the values observed at the regional background sites Mauna Loa and Waliguan.


Page 21, lines 577-579 “Overall, the .... fire etc.” Importantly it shares the transport mechanisms.
We would like to thank reviewer for raising the role of transport mechanism in this section and it has been added in lines 647-650. The revised sentence reads as follows:

Overall, the positive and high correlations between CH₄ and CO mixing ratios and between CH₄ and CO₂ in the pre-monsoon and winter indicate common sources, most likely combustion related sources such as vehicular emission, brick kilns, agriculture fire etc., or the same source regions (i.e. their transport from outside the Kathmandu Valley due to regional atmospheric transport mechanisms).

Pages 21-22, Sec. 3.5 I strongly recommend authors to remove the whole section. It is not at all straightforward, as assumed here, to determine the impact of emission sources and transport, based on concentration measurements from single site. It does not make any sense unless a dedicated further study is involved to justify the stated assumptions here. The section, as in the present shape, does not meet scientific reasoning; hence need to be removed.

We are in agreement with the reviewer that without any good supporting evidence, it can be difficult to accept the assumptions made. Thus, as suggested, the whole section has removed from the manuscript and the remaining sections are renumbered.

Page 23, lines 633-641 “Based on the .... post-monsoon seasons” This could be a likely scenario. Have these interpretations been supported by any emission inventories available? It is also important to point out the associated uncertainties involved in separating different emission sectors based on this approach. Note that this approach cannot separate near and far field sources, different lifetimes of tracers etc.

Yes, we agree that the source identification based on CO/CO₂ ratio is indicative, not a definitive evidence. We apologize if we sounded definitive in our sentences and conclusion derived from the ratio analysis. We have also stated clearly in the paragraph that this method or the values associated with the source (Table 5) or our estimated values may have large uncertainty. A few additional lines have been added 680-682 to convey our cautious approach. Although we were unable to estimate the standard deviation of the ratio in Table 5, we have included the standard
deviation of our calculated ratio in Table 6, and added useful statistics such as geometric mean and geometric standard deviation and their upper and lower bounds. The whole paragraph is significantly modified, and as supporting evidence, we also have added emission source sectors in the Kathmandu Valley based on a high resolution emission inventory (Sadavarte et al., 2017, in preparation). Here is the modified paragraph (lines 715-730):

Although ratio of CO/CO₂ is a weak indicator of sources and the mean ratio has large variance (See Table 6), the conclusions drawn from using Figure 8 and the above mentioned classification are not conclusive. The estimated CO/CO₂ ratio tentatively indicates that the local plume impacting the measurement site (Bode) from the north and east could be residential and/or diesel combustion. The estimated CO/CO₂ ratio of the local plume from the south and west generally falls in the 15-45 range, which could indicate emissions from brick kilns and inefficient gasoline vehicles. Very high ratios were also estimated from the south west during the post-monsoon season. Among other possible sources, this may indicate agro-residue open burning.

The emission inventory for CO identifies (aggregate for a year) residential, and gasoline related emission from transport sector (Sadavarte et al., 2017, in preparation). The inventory is not yet temporally resolved, so no conclusion can be drawn about the sources with respect to different seasons. From the 1km x1km emission inventory of the Kathmandu Valley for 2011, the estimated sectoral source apportionment of CO is residential (37%), transport sector (40%) and industrial (20%). The largest fraction from the residential sector is cooking (24%) whereas the majority of transport sector related CO in the Kathmandu Valley is from gasoline vehicles.

Reference:


Page 25, lines 704-706 “but it is clear that” What makes it clear?

We have removed the confusing words and rephrased the sentence in lines 793-795 as:
The cause of this diurnal pattern at Chanban is presently unclear, but the levels could be representative of the regional background throughout the day and show only limited influences of local emissions.

Page 26, lines 731-732 “Regional transport .... during pre-monsoon” I don’t see any valid justification for this throughout.

The sentence has been removed from the conclusion section because we also realized that there is not a justifiable connection with the result presented in the manuscript.

Page 27, lines 743-744 “Low values .... mixing ratios.” Please provide supporting details.

We have rephrased the sentence (lines 832-833):

Low values of CH₄ and CO₂ mixing ratios at the Chanban site could represent regional background mixing ratios.

Page 27, line 746 “useful for evaluation of satellite measurements and climate” How?

We have included the reason how the measurement is useful for satellite evaluation and rephrased the sentence to make it clear (lines 835-837):

These observations can be useful as ground-truthing for evaluation of satellite measurements, as well as climate and regional air quality models.

Page 27, lines 747-749 “The analysis ... Kathmandu Valley”, Please, remove this sentence. Note that this is not met here and the study only demonstrates the observational variations of GHGs in the study region.
We agree with the reviewer that this study focuses on observational variations of mainly CH$_4$ and CO$_2$. However, this study along with recent studies will help in addressing mitigation of the pollution in the study region. Thus, we have rephrased the sentence in lines 837-839 as:

The **overall** analysis presented in the paper **will contribute along with other recent measurements and analysis to** providing a sound scientific basis for reducing emissions of greenhouse gases and air pollutants in the Kathmandu Valley.

Reference Please check. Formatting issues and sometimes journal details (or other important parts) are missing

We have re-checked the references and tried to resolve formatting and other issues in the section.

Table 4 Columns 4 & 5: What are these values? Looks like monthly values for Bode during Aug., and Sep. 2013; but then why are they different from corresponding 2$^{nd}$ column of the table 4?

These are monthly values for Bode, which were simultaneously measured with another site at Chanban during August and September 2015. The information has been included in Table 4.

Table 6 Column 2: This “*” meant for?

We would like to thank reviewer for noticing this symbol. The meaning of the symbol has been included with a note at the bottom of the Table 6 as:

*The morning peak was one hour delayed in winter, thus the 8:00-10:00 period data was used in the analysis.*

Figure 4 I didn’t follow the fig. well. What I understood is that the plot shows the frequency of hourly mixing ratios w.r.t the frequency of prevailing wind direction. Did it also take into account the wind speed? Then what about different percentages shown? For example, does it
mean that 5% of sample time in August, the wind was from NE and “CH4_corrected” is above 2.5 ppm in which less than 1% time (in my eyes), CH4 is in 3-3.3 ppm range? In that case, it’s statistically difficult to say that the monthly enhancement is due to the polluted air masses from the NE and E. By the way, what are these “_corrected” values for CH4 and CO2? What about March-April scenario for CO2_corrected? Did two plots (Figs.2 and 3) use same set of master data, or any quality filtering had been done other than monthly averaging?

We are sorry for creating confusion by keeping “CH4_corrected” in the legend of Figure 4. CO2 and CH4 corrected means water corrected values of them, which we explained in section 2.2….. The CH4 and CO2 data used in the whole analysis (including Figure 2 and 3) are the same and they are water corrected values. We did not use any additional filtering while making these plots in the analysis. To avoid confusion, we have replaced CH4_corrected and CO2_corrected by CH4 and CO2 respectively in the legend of Figure 4.
Seasonal and diurnal variations of methane and carbon dioxide in the Kathmandu Valley in the foothills of the central Himalaya

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Abstract

The SusKat-ABC (Sustainable Atmosphere for the Kathmandu Valley- Atmospheric Brown Clouds) international air pollution measurement campaign was carried out during December 2012-June 2013 in the Kathmandu Valley and surrounding regions in Nepal. The Kathmandu Valley is a bowl-shaped basin with a severe air pollution problem. This paper reports measurements of two major greenhouse gases (GHGs), methane (CH\textsubscript{4}) and carbon dioxide (CO\textsubscript{2}), along with the pollutant CO, that began during the campaign and were extended for a year at the SusKat-ABC’s supersite in Bode, a semi-urban location in the Kathmandu Valley. Simultaneous measurements were also made during 2015 in Bode and at a nearby rural site (Chanban), ~25 km (aerial distance) to the southwest of Bode, on the other side of a tall ridge. The ambient mixing ratios of methane (CH\textsubscript{4}), carbon dioxide (CO\textsubscript{2}), water vapor, and carbon monoxide (CO) were measured with a cavity ring down spectrometer (Picarro G2401, USA),
Simultaneous measurements were also made at Chanban from 15 July to 3 October 2015. These measurements are the first of their kind in the central Himalayan foothills. At Bode, the annual average mixing ratios of CO₂ and CH₄ were 419.34(±236.09) ppm and 2.1923(±0.066224) ppm, respectively. These values are higher than the levels observed at background sites such as Mauna Loa, USA (CO₂: 396.8 ± 2.0 ppm, CH₄: 1.831 ± 0.110 ppm) and Waliguan, China (CO₂: 397.7 ± 3.6 ppm, CH₄: 1.879 ± 0.009 ppm) during the same period, and at other urban/semi-urban sites in the region such as Ahmedabad and Shadnagar (India) and Nanjing (China). They varied slightly across the seasons at Bode, with seasonal average CH₄ mixing ratios being 2.157(±0.230) ppm in the pre-monsoon season, 2.199(±0.241) ppm in the monsoon, 2.210(±0.200) ppm in the post-monsoon, and 2.214(±0.209) ppm in the winter season. The average CO₂ mixing ratios were 426.2(±25.5) ppm in pre-monsoon, 413.5(±24.2) ppm in monsoon, 417.3(±23.1) ppm in post-monsoon, and 421.9(±20.3) ppm in winter season. The maximum seasonal mean mixing ratio of CH₄ in winter was only 0.057 ppm or 2.6% higher than the seasonal minimum during the pre-monsoon period, while CO₂ was 12.8 ppm or 3.1% higher during the pre-monsoon period (seasonal maximum) than during the monsoon (seasonal minimum). On the other hand, the CO mixing ratio at Bode was 191% higher during the winter than during the monsoon season. The enhancement in CO₂ mixing ratios during the pre-monsoon season is associated with additional CO₂ emissions from forest fire and agro-residue burning in northern South Asia in addition to local emissions in the Kathmandu Valley. Published CO/CO₂ ratios of different emission sources in Nepal and India were compared with the observed CO/CO₂ ratios in this study. This comparison indicated that the major sources in the Kathmandu Valley were residential cooking and vehicle exhaust in all seasons except winter. In winter, the brick kiln emissions were a major source. Simultaneous measurement in Bode and Chanban (15 July-3 Oct 2015) revealed that the mixing ratio of CO₂, CH₄ and CO mixing ratios were 3.8%, 12%, and 64% higher in Bode than Chanban. Kathmandu Valley, thus, has significant emissions from local sources, which can also be attributed to its bowl shaped geography that is conducive to pollution build-up. At Bode, all three gas species (CO₂, CH₄ and CO) showed strong diurnal patterns in their mixing ratios with a pronounced morning peak (ca. 08:00), a dip in the afternoon, and again gradual
increase through the night until the next morning, whereas CH₄ and CO at Chanban did not show any noticeable diurnal variations.

These measurements provide the first insights into diurnal and seasonal variation of key greenhouse gases and air pollutants and their local and regional sources, which are important information for the atmospheric research in the region.

1 Introduction

The average atmospheric mixing ratios of two major greenhouse gases (GHGs), CO₂ and CH₄, have increased by about 40% (from 278 to 390.5 ppm) and about 150% (from 722 to 1803 ppb) respectively since pre-industrial times (~1750 AD). This is mostly attributed to anthropogenic emissions (IPCC, 2013). The current global annual rate of increase of the atmospheric CO₂ mixing ratio is 1-3 ppm, with average annual mixing ratios now exceeding a value of 400 ppm at the background reference location in Mauna Loa (WMO, 2016). Between 1750 and 2011, 240555±1085 PgC of anthropogenic CO₂ was accumulated in the atmosphere of which two thirds were contributed by fossil fuel combustion and cement production, with the remaining coming from deforestation and land use/land cover changes (IPCC, 2013). CH₄ is the second largest gaseous contributor to anthropogenic radiative forcing after CO₂ (Forster et al., 2007). The major anthropogenic sources of atmospheric CH₄ are rice paddies, ruminants and fossil fuel use, contributing approximately 60% to the global CH₄ budget (Chen and Prinn, 2006; Schneising et al., 2009). The remaining fraction is contributed by biogenic sources such as wetlands and fermentation of organic matter by microbes in anaerobic conditions (Conrad, 1996).

Increasing atmospheric mixing ratios of CO₂ and CH₄ and other GHGs and short-lived climate-forcing pollutants (SLCPs) such as black carbon (BC) and tropospheric ozone (O₃) have caused the global mean surface temperature to increase by 0.85°C from 1880 to 2012. The surface temperature is expected to increase further by up to 2 degrees at the end of the 21st century in most representative concentration pathways (RCP) emission scenarios (IPCC, 2013). The increase in surface temperature is linked to melting of glaciers and ice sheets, sea level rise,
extreme weather events, loss of biodiversity, reduced crop productivity, and economic losses (Fowler and Hennessy, 1995; Guoxin and Shibasaki, 2003).

Seventy percent of global anthropogenic CO$_2$ is emitted in urban areas (Fragkias et al., 2013). Developing countries may have lower per capita GHG emissions than developed countries, but the large cities in developing countries, with their high population and industrial densities, are major consumers of fossil fuels and thus, emitters of GHGs. South Asia, a highly populated region with rapid growth in urbanization, motorization, and industrialization in recent decades, has an ever increasing fossil fuel demand and its combustion emitted 444 Tg C/year in 2000 (Patra, et al., 2013), or about 5% of the global total CO$_2$ emissions. Furthermore, a major segment of the population in South Asia has an agrarian economy and uses biofuel for cooking activities, and agro-residue burning is also common practice in the region, which are important major sources of air pollutants and greenhouse gases in the region (CBS, 2011; Pandey et al., 2014; Sinha et al., 2014).

The emission and uptake of CO$_2$ and CH$_4$ follow a distinct cycle in South Asia. By using inverse modeling, Patra et al. (2011) found also showed that a net CO$_2$ uptake (0.37 ± 0.20 Pg C yr$^{-1}$) during 2008 in South Asia and the uptake (sink) is highest peaks during July-September. And The remaining months acts as a weak gross sink but a moderate gross source for CO$_2$ in the region August, using an inversion constrained by regional measurements from commercial aircraft. The observed trend variation is linked with the growing seasons. Agriculture is a major contributor of methane emission. For instance, in India it contributes to 75% of CH$_4$ emissions (MoEF, 2007). Ambient CH$_4$ concentrations are highest during June to September (peaking in September) in South Asia which are also the growing months for rice paddies (Goroshi et al., 2011). The minimum column averaged ambient CH$_4$ concentrations mixing ratios are observed in February-March (Prasad et al., 2014).

Climate change has impacted South Asia in several ways, as evident in temperature increase, change in precipitation patterns, higher incidence of extreme weather events (floods, droughts, heat waves, cold waves), melting of snowfields and glaciers in the mountain regions, and impacts on ecosystems and livelihoods (ICIMOD, 2009; MoE, 2011). Countries such as Nepal
are vulnerable to impacts of climate change due to inadequate preparedness for adaptation to impacts of climate change (MoE, 2011). Decarbonization of its economy can be an important policy measure in mitigating climate change. Kathmandu Valley is one of the largest metropolitan cities in the foothills of the Hindu Kush-Himalaya which has significant reliance on fossil fuels and biofuels. In 2005, fossil fuel burning accounted for 53% of total energy consumption in the Kathmandu Valley, while biomass and hydroelectricity were 38% and 9%, respectively (Shrestha and Rajbhandari, 2010). Fossil fuel consumed in the Kathmandu Valley accounts for 32% of the country’s fossil fuel imports, and the major fossil fuel consumers are residential (53.17%), transport (20.80%), industrial (16.84%), and commercial (9.11%) sectors. Combustion of these fuels in traditional technologies such as Fixed Chimney Bulls Trench Kiln (FCBTK) and low efficiency engines (vehicles, captive power generator sets etc.) emit significant amounts of greenhouse gases and air pollutants. This has contributed to elevated ambient concentrations of particulate matter (PM), including black carbon and organic carbon, and several gaseous species such as ozone, polycyclic aromatic hydrocarbons (PAHs), acetonitrile, benzene and isocyanic acid (Pudasainee et al., 2006; Aryal et al., 2009; Panday and Prinn, 2009; Sharma et al., 2012; World Bank, 2014; Chen et al., 2015; Putero et al., 2015; Sarkar et al., 2016). The ambient levels often exceed national air quality guidelines (Pudasainee et al., 2006; Aryal et al., 2009; Putero et al., 2015) and are comparable or higher than ambient levels observed in other major cities in South Asia.

Past studies in the Kathmandu Valley have focused mainly on a few aerosols species (BC, PM) and short-lived gaseous pollutants such as ozone and carbon monoxide (Pudasainee et al., 2006; Aryal et al., 2009; Panday and Prinn, 2009; Sharma et al., 2012, Putero et al., 2015). To the best of authors’ knowledge, no direct measurements of CO₂ and CH₄ are available for the Kathmandu Valley. Recently, emission estimates of CO₂ and CH₄ were derived for the Kathmandu Valley using the International Vehicle Emission (IVE) model (Shrestha et al., 2013). The study estimated 1554 Gg of annual emission of CO₂ from a fleet of vehicles (that consisted of public buses, 3-wheelers, taxis and motor cycles; private cars, trucks and non-road vehicles were not included in the study) for the year 2010. In addition, the study also estimated 1.261 Gg of CH₄ emitted from 3 wheelers (10.6 %), taxis (17.7 %) and motorcycles (71 %) for 2010.
This study presents the first 12 months of measurements of two key GHGs, CH$_4$ and CO$_2$ along with other trace gases and meteorological parameters in Bode, a semi-urban site in the eastern part of the Kathmandu Valley. The year-long measurement in Bode is a part of the SusKat-ABC (Sustainable Atmosphere for the Kathmandu Valley – Atmospheric Brown Clouds) international air pollution measurement campaign conducted in and around the Kathmandu Valley from December 2012 to June 2013. Details of the SusKat-ABC campaign are described in Rupakheti et al. (2017, manuscript in preparation). The present study provides a detailed account of seasonal and diurnal behaviors of CO$_2$ and CH$_4$ and their possible sources. To examine the rural-urban differences and estimate the urban enhancement, these gaseous species were also simultaneously measured for about three months (Jul-Oct) in 2015 at Chanban, a rural site about 25 km (aerial distance) outside and southwest of Kathmandu Valley. The seasonality of the trace gases and influence of potential sources in various (wind) directions are further explored by via ratio analysis. This measurement provides unique data from highly polluted but relatively poorly studied region (central Himalayan foothills in South Asia) which could be useful for validation of emissions estimates, model outputs and satellite observations. The study, which provides new insights on potential sources, can also be a good basis for designing mitigation measures for reducing emissions of air pollutants and controlling greenhouse gases in the Kathmandu Valley and the region.

2 Experiment and Methodology

2.1 Kathmandu Valley

The Kathmandu Valley consists of three administrative districts: Kathmandu, Lalitpur, and Bhaktapur, situated between 27.625° N, 27.75° N and 85.25°E, 85.375°E. It is a nearly circular bowl-shaped valley with a valley floor area of approximately 340 km$^2$ located at an altitude of 1300 m mean sea level (masl). The surrounding mountains are close to 2000-2800 in height above sea level with five mountain passes located at about 200-600 m above the valley floor and an outlet for the Bagmati River southwest of the Kathmandu Valley. Lack of decentralization in Nepal has resulted in the concentration of economic activities, health and education facilities, the service sector, as well as most of the central governmental offices in the Kathmandu Valley.
Consequently, it is one of the fastest growing metropolitan areas in South Asia with a current population of about 2.5 million, and the population growth rate of 4% per year (World Bank, 2013). Likewise, approximately 50% of the total vehicle fleet (2.33 million) of the country is in Kathmandu Valley (DoTM, 2015). The consumption of fossil fuels such as liquefied petroleum gas (LPG), kerosene for cooking and heating dominates the residential consumption, while the rest use biofuel (fuelwood, agro-residue, animal dung) for cooking and heating in the Kathmandu Valley. The commercial sector is also growing in the valley, and the latest data indicate the presence of 633 industries of various sizes. These are mainly associated with dyeing, brick kilns, and manufacturing industries. Fossil fuels such as coal and biofuels are the major fuels used in brick kilns. Brick kilns are reported as one of the major contributors of air pollution in the Kathmandu Valley (Chen et al., 2015; Kim et al., 2015; Sarkar et al., 2016). There are about 115 brick industries in the valley (personal communication with M. Chitrakar, President of the Federation of Nepalese Brick Industries). Acute power shortage in the Valley is common all around the year, especially in the dry season (winter/pre-monsoon) when the power cuts can last up to 12 hours a day (NEA, 2014). Energy demand during the power cut period is met with the use of small (67% of 776 generators surveyed for the World Bank study was with capacity less than 50kVA) but numerous captive power generators (diesel/petrol), which further contribute to valley’s poor air quality. According to the World Bank’s estimate, over 250,000 such generator sets are used in the Kathmandu Valley alone, producing nearly 200 MW of captive power, and providing about 28% of the total electricity consumption of the valley (World Bank, 2014). Apart from these sources, trash burning, which is a common practice (more prevalent in winter) throughout the valley, is one of the major sources of air pollutants and GHGs.

Climatologically, Kathmandu Valley has a sub-tropical climate with annual mean temperature of 18°C, and annual average rainfall of 1400 mm, of which 90% occurs in monsoon season (June-September). The rest of the year is dry with some sporadic rain events. The wind circulation at large scale in the region is governed by the Asian monsoon circulation and hence the seasons are also classified based on such large scale circulations and precipitation: Pre-Monsoon (March-May), Monsoon (June-September), Post-Monsoon (October-November) and Winter (December-February). Sharma et al. (2012) used the same classification of seasons while explaining the
seasonal variation of BC concentrations observed in the Kathmandu Valley. Locally in the valley, the mountain-valley wind circulations play an important role in influencing air quality.

The wind speed at the valley floor is calm ($\leq 1$ m s$^{-1}$) in the morning and night, while a westerly wind develops after 11:00 AM in the morning till dusk, and switches to a mild easterly at night (Panday and Prinn, 2009; Regmi et al., 2003). This is highly conducive to building up of air pollution in the valley, which gets worse during the dry season.

2.2 Study sites

Two sites, a semi-urban site within the Kathmandu Valley and a rural site outside the Kathmandu Valley, were selected for this study. The details of the measurements carried out in these sites is described Table 1 and in section 2.2.1 and 2.2.2.

2.2.1 Bode (SusKat-ABC supersite)

The SusKat-ABC supersite was set up at Bode, a semi-urban location (Figure 1) of the Madhyapur Thimi municipality in the Bhaktapur district in the eastern side of the Kathmandu Valley. The site is located at 27.68°N latitude, 85.38°E longitude, and 1344 masl. The local area around the site has a number of scattered houses and agricultural fields. The agriculture fields are used for growing rice paddies in the monsoon season. It also receives outflow of polluted air from three major cities in the valley: Kathmandu Metropolitan City and Lalitpur Sub-metropolitan City, both mainly during daytime, and Bhaktapur Sub-metropolitan City mainly during nighttime. Among other local sources around the site, about 10 brick kilns are located in the east and southeast direction, approximately within 1-4 km from the site which are operational only during dry season (January to April). There are close to 20 small and medium industries (pharmaceuticals, plastics, electronics, tin, wood, aluminum, iron, and fabrics etc.) scattered in the same direction. The Tribhuvan International Airport (TIA) is located approximately 4 km away to the west of the Bode site.

2.2.2 Chanban

Chanban is a rural/background site in Makwanpur district outside of the Kathmandu Valley (Figure 1). This site is located ~25 km aerial distance due southwest from Bode. The site is
located on a small ridge (27.65° N, 85.14° E, 1896 masl) between two villages - Chitlang and Bajrabarahi - within the forested watershed area of Kulekhani Reservoir, which is located ~ 4.5 km southwest of the site. The instruments were set up on the roof of 1-storey building in an open space inside the Nepali Army barrack. There was a kitchen of the army barrack at about 100 m to the southeast of the measurement site. The kitchen uses LPG, electricity, kerosene, and firewood for cooking activities.

2.3 Instrumentation

The measurements were carried out in two phases in 2013-2014 and 2015. In phase one, a cavity ring down spectrometer (Picarro G2401, USA) was deployed in Bode to measure ambient CO₂, CH₄, CO, and water vapor mixing ratios. Twelve months (6 March 2013 - 5 March 2014) of continuous measurements were made in Bode. The operational details of the instruments deployed in Bode are also provided in Table 1. In phase two, simultaneous measurements were made in Bode and Chanban for a little less than 3 months (15 July to 03 October 2015).

The Picarro G2401 analyzer quantifies spectral features of gas phase molecules by using a novel wavelength-scanned cavity ring down spectroscopic technique (CRDS). The instrument has a 30 km path length in a compact cavity that results high precision and sensitivity. Because of the high precision wavelength monitor, it uses absolute spectral position and maintains accurate peak quantification. Further, it only monitors the special features of interest for reducing the drift. The instrument also has water correction to report dry gas fraction. The reported measurement precision for CO₂, CH₄, CO and water vapor in dry gas is < 150 ppb, < 30 ppb, < 1 ppb and < 200 ppm for 5 seconds with 1 standard deviation (Picarro, 2015).

In Bode, the Picarro analyzer was placed on the 4th floor of a 5-storey building with an inlet at 0.5 m above the roof of the building with a 360 degree view (total inlet height: 20 m above ground). The sample air was filtered at the inlet to keep dust and insects out and was drawn into the instrument through a 9 m Teflon tube (1/4 inches ID). The Picarro analyzer was set to record data in every 5 second and recorded both directly sampled data and water corrected data of CO₂ and CH₄. In this paper, only water-corrected or dry mixing ratios of CH₄ and CO₂ were used to calculate the hourly averages for diurnal and seasonal analysis.
The instruments were factory calibrated before commencing the field measurement. Picarro G2401 model is designed for remote application and long term deployment with minimal drift and less requirement for intensive calibration (Crosson, 2008) and thus was chosen for the current study in places like Kathmandu where there is no or limited availability of high quality reference gases. Regular calibration of Picarro G2401 in field during 2013-2014 deployment was not conducted due to challenges associated with the quality of the reference gas, especially for CO and CH₄. One time calibration was performed for CO₂ (at 395, and 895 ppmv) in July 2015 before commencing the simultaneous measurement in Bode and Chanban in 2015. The % difference between CO₂ mixing ratio reported by the analyzer and the differed by approximately 5% at reference mixing ratio was within 5%. CO observations from Picarro G2401 were compared with observations from another CO analyzer (Horiba, model AP370) that was also operated in Bode for 3 months (March - May 2013). Horiba CO monitor was a new unit, which was factory calibrated before its first deployment in Bode. Nevertheless, this instrument was inter-compared with another CO analyzer (same model) from the same manufacturer prior to the campaign and its correlation coefficient was 0.9 [slope of data from the new unit (y-axis) vs the old unit (x-axis) = 1.09]. Primary gas cylinders from Linde UK (1150 ppbv) and secondary gases from Ultra-Pure Gases and Chemotron Science Laboratories (1790 ppbv) were used for the calibration of CO instrument. Further details on CO measurements and calibration of Horiba AP370 can be found in Sarangi et al. (2014; 2016). Statistically significant correlation (r = 0.99, slope = 0.96) was found between Picarro and Horiba hourly average CO mixing ratio data (Supplementary Information Figure S1). Furthermore, the monthly mean difference between these two instruments (Horiba AP370 minus Picarro G2401) was calculated to be 0.02 ppm (3%), 0.04 ppm (5%) and 0.02 ppm (4%) in March, April and May, respectively. For the comparison period of 3 months, the mean difference was 0.02 ppm (4%). Overall differences were small to negligible during the comparison period and thus, adjustment in the data was deemed not necessary.

Besides highly selective to individual species, Picarro G2401 has a water correction function and thus accounts for the any likely drift in CO, CO₂ and CH₄ mixing ratios with the fluctuating water vapor concentration (Chen et al., 2013; Crosson, 2008). Crosson (2008) also estimated a
peak to peak drift of 0.25 ppmv. Further, Crosson (2008) observed a 1.2 ppbv/day drift in CO₂ after 170 days from the initial calibration. For a duration of one year the drift will be less than 1 ppmv, which is less than 1% of the observed mixing ratio in (hourly ranges: 376-537 ppm) Bode even if the drift was in same magnitude as in case of Crosson (2008). Crosson (2008) reported 0.8 ppbv peak to peak drift in CH₄ measurements for 18 days after the initial calibration.

There were other instruments concurrently operated in Bode; a ceilometer for measuring mixing layer height (Vaisala Ceilometer CL31, Finland), and an Automatic Weather Station (AWS) (Campbell Scientific, USA). The ceilometer was installed on the rooftop (20 m above ground) of the building (Mues et al., 2017). For measuring the meteorological parameters, a Campbell Scientific AWS (USA) was set up on the roof of the building with sensors mounted at 2.9 m above the surface of the roof (22.9 m from the ground). The Campbell Scientific AWS measured wind speed and direction, temperature, relative humidity and solar radiation every minute. Temperature and rainfall data were taken from an AWS operated by the Department of Hydrology and Meteorology (DHM), Nepal at the Tribhuvan International Airport (TIA, see Figure 1), ~4 km due west of Bode site.

At Chanban, the inlet for Picarro gas analyzer was kept on the rooftop ~3 m above the ground and the sample air was drawn through a 3 m long Teflon tube (1/4 inches ID). The sample was filtered at the inlet with a filter (5-6 µm pore size) to prevent aerosol particles from getting into the analyzer. An automatic weather station (Davis Vantage Pro2, USA) was also set up in an open area, about 17 m away from the building and with the sensors mounted at 2 m above ground.

3. Results and discussion

The results and discussions are organized as follow: Sub-section 3.1 describes a year round variation in CH₄, CO₂, CO and water vapor at Bode; sub-sections 3.2, 3.3 and 3.4 present the analysis of the observed diurnal, monthly and seasonal variations and diurnal variation. Sub-sections 3.4 and 3.5, 3.6, 3.7 discusses the impact of city pollution at the measurement site at Bode, influence of regional pollution and transport about the interrelation of CO₂, CH₄ and CO
and potential emission sources in the valley and sub-section 3.68 compares and contrasts CH₄, CO₂, CO at Bode and Chanban.

3.1 Time series of CH₄, CO₂, CO and water vapor mixing ratios

Figure 2 shows the time series of hourly mixing ratios of CH₄, CO₂, CO, and water vapor at Bode. Meteorological data from Bode and the Tribhuvan International Airport are also shown in Figure 2. Data gaps in Figure 2a and 2b were due to maintenance of the measurement station. In general, the changes observed in CO mixing ratio was higher in terms of % change than the variations observed in CH₄ and CO₂ mixing ratios during the sampling period. In contrast, CO mixing ratios decreased and water vapor mixing ratios increased significantly during the rainy season (June-September). For the entire sampling period, the annual average (± one standard deviation) of CH₄, CO₂, CO, and water vapor mixing ratios were 2.1923 (±0.066-0.224) ppm, 419.34 (±6.0-23.9) ppm, 0.50 (±0.23-0.35) ppm, and 1.731 (±0.66-0.71)%, respectively. The relative variabilities for the annual average of CH₄, CO₂ and CO were thus 3%, 1.4% and 46%, respectively. Their variabilities at Mauna Loa were CH₄: 6% and CO₂: 0.5% and at Waliguan were CH₄: 0.48%, CO₂: 0.9%. The high variability in the annual mean, notably for CO in Bode could be indicative of the seasonality of emission sources and meteorology. The annual CH₄ and CO₂ mixing ratios were compared to the historical background site (Mauna Loa Observatory, Hawaii, USA) and the background site (Waliguan, China) in Asia, which will provide insight on spatial differences. The selection of neighboring countries’ (i.e., Indian and China’s) urban and semi-urban sites, where many emission sources are typical for the region, for comparison provides information on relative differences (higher/lower), which will help in investigating possible local emission sources in the valley. As expected, annual mean of CH₄ and CO₂ mixing ratios in the Kathmandu Valley were higher than the levels observed at background sites in the region and elsewhere for the same period (Table 4). We performed a significance test at 95% confidence level (t-test) of the annual mean values between the sites to evaluate whether the observed difference is statistically significant (p < 0.05), which was confirmed for the. We found the difference in annual mean CH₄ and CO₂ between Bode and Mauna Loa, and between Bode and Waliguan were statistically significant. CH₄ was nearly 230% higher at Bode than at Mauna Loa observatory (1.831 ± 0.110 ppm) (Dlugokencky et al., 20176) and ca.17% higher than at Mt.
Waliguan (1.879 ± 0.009 ppm) in China for the same observation period (Dlugokencky et al., 2016). The slightly higher small difference CH$_4$ mixing ratios between at Bode and Waliguan in comparison to than at Mauna Loa Observatory could signal indicates the higher mixing ratio of CH$_4$ in these two Asian sites. The be due to rice farming as a key source of CH$_4$ in this part of Asia. Thus, it could be associated with such agricultural activities in this region. Similarly, the annual average CH$_4$ at Bode during 2013-14 was found comparable to higher than urban/semi-urban sites in India, such as an urban site in Ahmedabad (1.880 ± 0.4 ppm, i.e., variability: 21.3%) in India for 2002 (Sahu and Lal, 2006) and 14% higher than in Shadnagar (1.92 ± 0.07 ppm, i.e., variability: 3.6%), a semi-urban site in Telangana state (~70 km north from Hyderabad city) during 2014 (Sreenivas et al., 2016). Likewise, the difference between annual mean mixing ratios at Bode (419.3 ± 4.6 ppm, 1.4% variability) vs. Mauna Loa (396.8 ± 2.0 ppm, 0.5% variability) (NOAA, 2015) and Bode vs. Waliguan (397.7 ± 3.6 ppm, 0.9% variability) (Dlugokencky et al., 2016) is statistically significant (p <0.05). the annual average CO$_2$ mixing ratio at Bode (419.4 ±23.9 ppm) during the observation period was 5.7% higher than at Mauna Loa Observatory (396.76 ppm) (Tans and Keeling, 2014) and 5.5% higher than at Mt. Waliguan (397.7 ppm). The CO$_2$ mixing ratio in the Kathmandu Valley was also found to be higher than the levels observed in Shadnagar (394 ± 2.9 ppm) during 2014, Ahmedabad city (413 ± 13.7 ppm) in India during November 2013 to May 2015, and an urban site at Nanjing (406.5 ± 20 ppm) in China (Huang et al., 2015; Sreenivas et al., 2016; Chandra et al., 2016).

The high CH$_4$ and CO$_2$ mixing ratios at Bode in comparison to Ahmedabad and Shadnagar and Nanjing could be due to more than 115 coal-biomass fired brick kiln, some of them are located near the site (less than 4 km) and confinement of pollutants within the Valley due to bowl shaped topography of the Kathmandu Valley. Although Ahmedabad and Nanjing sites are in a big cityies with high population larger than Kathmandu Valley but the measurement sites are far from the nearby heavy polluting industries and situated in plains, where ventilation of pollutants would be more efficient as opposed to the Kathmandu Valley. The major polluting sources were industries, residential cooking and transport sector in Ahmedabad (Chandra et al., 2016). Anthropogenic emission, synoptic circulation, terrestrial biosphere had important role on CO$_2$ mixing ratios in Nanjing (Huang et al., 2015). Shadnagar is a small town with a population of
0.16 million and major sources were industries (small-medium), biomass burning in residential cooking (Sreenivas et al., 2016).

The monthly average of CO₂ mixing ratios in 2015 in Chanban (Aug: 403.4, Sep: 399.1 ppm) were slightly higher than the background sites at Mauna Loa Observatory (Aug: 398.89 ppm, Sep: 397.63 ppm) (NOAA, 2015) and Mt. Waliguan (Aug: 394.55 ppm, Sep: 397.68 ppm) (Dlugokencky et al., 2016a). For these two months in 2015, CH₄ mixing ratios were also higher in Bode (Aug: 2.281.11 ppmb, Sep: 2.371.09 ppmb) and Chanban (Aug: 2.05049.71 ppmb, Sep: 2.1024.75 ppmb) compared to Mauna Loa Observatory (Aug: 1.831.04 ppmb, Sep: 1.8465.68 ppmb) (Dlugokencky et al., 20176)) and Mt. Waliguan (Aug: 1.9154.99–ppmb, 1.911.21 ppmb) (Dlugokencky et al., 2016). The low differences in CO₂ between Chanban and background sites mentioned above indicate the less number of and/or less intense CO₂ sources at Chanban during these months because of the lack of burning activities due to rainfall in the region. The garbage and agro-residue burning activities were also absent or reduced around Bode during the monsoon period. However, high CH₄ values in August and September in Bode, Chanban and Mt. Waliguan in comparison to Mauna Loa Observatory may indicate the influence of CH₄ emission from paddy fields in the Asian region.

3.2 Monthly and Seasonal variations

Figure 3 shows the monthly box plot of hourly CH₄, CO₂, CO and water vapor observed for a year in Bode. Monthly and seasonal averages of CH₄ and CO₂ mixing ratios at Bode are summarized in Table 2 and 3. CH₄ were lowest during May-July (ranges from 2.093-2.129 ppm) period and highest during August-September (2.274-2.301 ppm), followed by winter. In addition to the influence of active local sources, the shallow boundary layer in winter was linked to elevated concentrations (Panday and Prinn, 2009; Putero et al., 2015, Mues et al., 20176). The low CH₄ values from May to July may be associated with the absence of brick kiln and frequent rainfall in these months. Brick kiln were operational during January to April. Rainfall also leads to suppression of open burning activities in the valley (see Figure 2b). The CH₄ was slightly higher (statistically significant, p<0.05) in monsoon season (July –September) than pre-monsoon season (unlike CO₂ which was higher in pre-monsoon), and could be associated with the addition.
of CH$_4$ flux from the water-logged rice paddies (Goroshi et al., 2011). There was a visible drop in CH$_4$ from September to October but remained consistently over 2.183 ppm from October to April with little variation between these months. Rice-growing activities are minimal or none in October and beyond, and thus may be related to the observed dip in CH$_4$ mixing ratio.

Comparison of seasonal average CH$_4$ mixing ratios at Bode and Shadnagar (a semi-urban site in India) indicated that CH$_4$ mixing ratios at Bode were higher in all seasons than at Shadnagar: pre-monsoon (1.89 ± 0.05 ppm), monsoon (1.85 ± 0.03 ppm), post-monsoon (2.02 ± 0.01 ppm), and winter (1.93 ± 0.05 ppm) (Sreenivas et al., 2016). The possible reason for lower CH$_4$ at Shadnagar in all season could be associated with geographical location and difference in local emission sources. The highest CH$_4$ mixing ratio in Shadnagar was reported in post-monsoon which was associated with harvesting in the Kharif season (July – October), while the minimum was in monsoon. Shadnagar is a relatively small city (population: ~0.16 million) compared to Kathmandu Valley and the major local sources which may have influence on CH$_4$ emission include bio-fuel, agro-residue burning and residential cooking.

The seasonal variation in CO$_2$ could be due to (i) generally reflects the seasonality of major emission sources such as brick kilns and regional emission sources such as vegetation fire and agriculture residue burn, (ii) seasonal growth of vegetation (CO$_2$ sink) (Patra et al., 2011) and (iii) atmospheric transport associated with regional synoptic atmospheric circulation (monsoon circulation and westerly disturbance in spring season) which could transport regional emission sources from vegetation fire and agriculture residue burning (Putero et al., 2015), and a local mountain-valley circulation effect (Kitada and Regmi, 2003; Panday et al., 2009). The concentrations of most pollutants in the region are lower during the monsoon period (Sharma et al., 2012, Marinoni, 2013; Putero et al., 2015) due to limited because frequent and heavy rainfall suppresses emission sources and partially due to rain washout. We saw a drop in the CO$_2$ mixing ratio during the rainfall period due to changes in various processes such as enhanced vertical mixing, uptake of CO$_2$ by vegetation and soils, and where relevant reduction in combustion sources. CO$_2$ can also dissolve into rainfall, forming carbonic acid, which may lead to a small decrease in the CO$_2$ mixing ratio as has been observed during heavy intensity rainfall (Mahesh et al., 2014; Chaudhari et al., 2007). Monsoon is also the growing season with higher CO$_2$
assimilation by plants than other seasons (Sreenivas et al., 2016). In contrast, winter, pre-
monsoon and post-monsoon season experiences an increase in emission activities in the
Kathmandu Valley (Putero et al., 2015).

The CO₂ mixing ratios were in the range of 376 - 537 ppm for the entire observation period.
Differences with CH₄ were observed in September and October where CO₂ was increasing
(mean/median) in contrast to CH₄ which showed the opposite trend. The observed increase in
CO₂ after October may be related to less or no rainfall, which results in the absence of rain-
washout and/or no suppression of active emission sources such as open burning activities.
However, the reduction in CH₄ after October could be due to reduced CH₄ emissions from paddy
fields, which were high in August-September. CO₂ remains relatively lower during July-August,
but it is over 420 ppm from January to May. Seasonal variation of CO₂ in Bode was similar in
seasonal variation but the values are higher than the values observed in Shadnagar, India
(Sreenivas et al., 2016).

The variations in CO were more distinct than CH₄ and CO₂ during the observation period
(Figure 3). The highest CO values were observed from January-April (0.71-0.91 ppm). The
seasonal mean of CO mixing ratios at Bode were: pre-monsoon (0.60 ±0.36 ppm), monsoon
(0.26±0.09 ppm), post-monsoon (0.40±0.15 ppm), and winter (0.76±0.43 ppm). The maximum
CO was observed in winter, unlike CO₂ which was maximum in pre-monsoon. The high CO in
winter was due to the presence of strong local pollution sources (Putero et al., 2015) and shallow
mixing layer heights. The addition of regional forest-fire and agro-residue burning augmented
CO₂ mixing ratios in pre-monsoon. The water vapor mixing ratio showed a seasonal pattern
opposite of CO, with a maximum in monsoon (2.53 %) and minimum in winter (0.95 %), and
intermediate values of 1.56 % in pre-monsoon and 1.55 % in post-monsoon season.

There were days in August-September when the CH₄ increases by more than 3 ppm (Figure 2).
Enhancement in CO₂ was also observed during the same time period. In the absence of tracer
model simulations, the directionality of the advected air masses is unclear. Figure 4 showed that
during these two months, it is likely that these high enhancements CO₂ mixing rations were
particularly high (> 450 CO₂ and > 2.5 ppm CH₄) with the air masses coming in from the
associated with the air mass from Northeast-East (NE-E) which had > 2.5 ppm CH₄ and > 450 ppm CO₂ (see Figure 4). CO during the same period was not enhanced and didn’t show any particular directionality compared to CH₄ and CO₂ (not shown in Figure 4c). Areas NE-E to Bode are predominantly irrigated (rice paddies) during August-September, and sources such as brick kilns were not operational during this time period. Goroshi et al. (2011) reported that June to September is a growing season for rice paddies in South Asia with high CH₄ emissions during these months and observed a peak in September in the atmospheric CH₄ column over India. Model analysis also points to high methane emissions in September which coincides with the growing period of rice paddies (Goroshi et al., 2011, Prasad et al., 2014). The CH₄ mixing ratios at Bode in January (2.233 ± 0.219 ppm) and July (2.129 ± 0.168 ppm) were slightly higher than the observation in Darjeeling (Jan: 1.929±0.056 ppm; Jul: 1.924±0.065 ppm), a hill station of eastern Himalaya (Ganesan et al., 2013). The higher CH₄ values in January and July at Bode compared to Darjeeling could be because of the influence of local sources, in addition to the shallow boundary layer in Kathmandu Valley. Trash burning and brick kilns are two major sources from December until April in the Kathmandu Valley while emission from paddy fields occurs during July-September in the Kathmandu Valley. In contrast, the measurement site in Darjeeling was located at higher altitude (2194 masl) and was less influenced by the local emission. The measurement in Darjeeling reflected a regional contribution. There are limited local source in Darjeeling such as wood biomass burning, natural gas related emission and vehicular emission (Ganesan et al., 2013).

The period between January and April had generally higher or the highest values of CO₂, CH₄ and CO at Bode. The measurement site was impacted mainly by local Westerly-Southwesterly winds (W-SW) and East-Southeast (E-SE). The W-SW typically has a wind speed in the range ~1 - 6 m s⁻¹ and was active during late morning to afternoon period (~11:00 to 17:00 NST, supplementary information Figure S2 and S3). Major cities in the valley such as Kathmandu Metropolitan City and Lalitpur Sub-metropolitan City are W-SW of Bode (Figure 1c). Wind from E-SE were generally calm (≤1m s⁻¹) and observed only during night and early morning hours (21:00 to 8:00 NST). The mixing ratio of all three species in air mass from the E-SE was significantly higher than in the air mass from W-SW (Figure 4). There are 10 biomass co-fired
brick kilns and Bhaktapur Industrial Estate located within 1-4 km E-SE from Bode (Sarkar et al., 2016). The brick kilns were only operational during January-April. Moreover, there were over 100 brick kilns operational in the Kathmandu Valley (Putero et al., 2015) which use low-grade lignite coal imported from India and biomass fuel to fire bricks in inefficient kilns (Brun, 2013).

Fresh emissions from main city center were transported to Bode during daytime by W-SW winds which mainly include vehicular emission. Compared to monsoon months (June-August), air mass from W-SW had higher values in all three species (Figure 4) during winter and pre-monsoon months. This may imply that in addition to vehicular emission, there are other potential sources which were exclusively active during these dry months. Municipal trash burning is also common in the Kathmandu Valley, with a reported higher frequency from December to February (Putero et al., 2015). The frequency in the use of captive power generator sets are highest during the same period, which is another potential source contributing to air coming from W-SW direction (World Bank, 2014; Putero et al., 2015).

Regional transport of pollutants into the Kathmandu Valley was reported by Putero et al. (2015). To relate the influence of synoptic circulation with the observed variability in BC and O₃ in the Kathmandu Valley, 5-day back trajectories (of air masses arriving in the Kathmandu Valley) were computed by Putero et al., (2015) using the HYSPLIT model. These individual trajectories which were initialized at 600 hPa, for the study period of one year and were clustered into nine clusters. Of the identified clusters, the most frequently observed clusters during the study period were the Regional and Westerly cluster or circulation (22% and 21%). The trajectories in the regional cluster originate within 10° x 10° around the Kathmandu Valley, whereas the majority of trajectories in this westerly cluster originated broadly around 20-40° N, ~60° E. Putero et al (2015) found that the regional and westerly synoptic circulation were favorable for high values of BC and O₃ in the Kathmandu Valley. Other sources of CO₂ and CH₄ could be due to vegetation fires which were also reported in the region surrounding the Kathmandu Valley during the pre-monsoon months (Putero et al., 2015). Similarly, high pollution events, peaking in the pre-monsoon, were observed at Nepal Climate Observatory-Pyramid (NCO-P) near Mt. Everest, which have been associated with vegetation fires in the Himalayan foothills and
northern IGP region (Putero et al., 2014). MODIS derived forest counts (Figure 5), which also
indicated high frequency of forest fire and farm fire from February to April and also during post-
monsoon season. It is interesting that the monthly mean CO₂ mixing ratio was maximum in April
(430 ± 27 ppm) which could be linked to the fire events. It is likely that the westerly winds
(>2.5-4.5 m s⁻¹) during the daytime (supplementary information Figure S2, S3) bring additional
CO₂ from vegetation fires and agro-residue burning in southern plains of Nepal including the
IGP region (Figure 5). Low values of CO₂ and CH₄ during June-July (Figure 3) was coincident
with the rainy season, and sources such as brick kiln emission, trash burning, captive power
generators, and regional agriculture residue burning and forest fires are weak or absent during
these months.

3.3 Diurnal Variation

Figure 6 shows the average seasonal diurnal patterns of CH₄, CO₂, CO, and water vapor mixing
ratios observed at Bode for four seasons. All the three gas species had a distinct diurnal pattern in
all seasons, characterized by maximum values in the morning hours (peaked around 7:00-9:00),
afternoon minima around 15:00-16:00, and a gradual increase through the evening until next
morning. There was no clear evening peak in CH₄ and CO₂ mixing ratios whereas CO shows an
evening peak around 20:00. The gradual increase of CO₂ and CH₄ in the evening in contrast to
the increase until evening peak traffic hours and later decay of CO may be indicative of a few
factors. As pointed out earlier, after the peak traffic hours, there are no particularly strong
sources of CO, especially in the monsoon and post-monsoon season. It is also likely that some of
the CO is decay due to nighttime katabatic winds which replace polluted air masses with cold
and fresh air from the nearby mountain (Panday and Prinn, 2009). As for the CO₂, the biosphere
respiration at night in the absence of photosynthesis can add additional CO₂ to the atmosphere
which especially in the very shallow nocturnal boundary layer may explains part of the increase
of the CO₂ mixing ratio. The well-defined morning and evening peaks observed in CO mixing
ratios are associated with the peaks in traffic and residential activities. The CH₄ and CO₂ showed
pronounced peaks in the morning hours (07:00-09:00) in all seasons with almost the same level
of seasonal average mixing ratios. CO had a prominent morning peak in winter and pre-monsoon
season, but the peak was significantly lower in monsoon and post-monsoon. The CO (~1-1.4
ppm) around 08:00-09:00 am in winter and pre-monsoon were nearly 3-4 times higher than in monsoon and post-monsoon season. It appears that CH$_4$ and CO$_2$ mixing ratios were continuously building up at night until the following morning peak in all seasons. The similar seasonal variations in CH$_4$ and CO$_2$ across all seasons could be due to their long-lived nature, as compared to CO, whose diurnal variations are strongly controlled by the evolution of the boundary layer. Kumar et al. (2015) also reported morning and evening peaks and an afternoon low in CO$_2$ mixing ratios in industrial, commercial, and residential sites in Chennai in India. The authors also found high early morning CO$_2$ mixing ratios at all sites and attributed it to the temperature inversion and stable atmospheric condition.

The daytime low CH$_4$ and CO$_2$ mixing ratios were due to (i) elevated mixing layer height in the afternoon (Figure 7), (ii) development of upslope wind circulation in the valley, and (iii) development of westerly and southwesterly winds which blows through the valley during the daytime from around 11 am to 5 pm (supplementary information Figure S2), all of which aid in dilution and ventilation of the pollutants out of the valley (Regmi et al., 2003; Kitada and Regmi, 2003; Panday and Prinn, 2009). In addition, the daytime CO$_2$ minimum in the summer monsoon is also associated with high photosynthetic activities in the valley as well as in the broader surrounding region. In the nighttime and early morning, the mixing layer height was low (only around 200-300 m in all seasons) and stable boundary layer for almost 17 hours a day. In the daytime it grows up to 800-1200 m for a short time (ca. from 11:00 to 6:00) (Mues et al., 20176, manuscript submitted to ACPD). Therefore the emissions from various activities in the evening after 18:00 (cooking and heating, vehicles, trash burning, and bricks factories in the night and morning) were trapped within the collapsing and shallow boundary layer, and hence mixing ratios were high during evening, night and morning hours. Furthermore, plant and soil respiration also increases CO$_2$ mixing ratio during the night (Chandra et al., 2016). However, Ganesan et al. (2013) found a distinct diurnal cycle of CH$_4$ mixing ratios with twin peaks in the morning (7:00-9:00), and afternoon (15:00-17:00) and a nighttime low in winter but no significant diurnal cycle in the summer of 2012 in Darjeeling, a hill station (2194 masl) in the eastern Himalaya. The authors described that the morning peaks could be due to the radiative heating of the ground in the morning, which breaks the inversion layer formed during night, and as a result, pollutants are
ventilated from the foothills up to the site. The late afternoon peaks match wind direction and wind speed (upslope winds) that could bring pollution from plains to mountains.

The diurnal variation of CO is also presented along with CO$_2$ and CH$_4$ in Figure 6c. CO is an indicator of primary air pollution. Although CO mixing ratio showed distinct diurnal pattern, it was different from the diurnal patterns of CO$_2$ and CH$_4$. CO diurnal variation showed distinct morning and evening peaks, afternoon minima, and a nighttime accumulation or decay. Nighttime accumulation in CO was observed only in winter and pre-monsoon and decay or decrease in monsoon season and post-monsoon season (Figure 7). The lifetime of CO (weeks to months) is very long compared to the ventilation timescales for the valley, so the different diurnal cycles would be due to differences in nighttime emissions. While the biosphere respires at night which may cause a notable increase in CO$_2$ in the shallow boundary layer, most CO sources (transport sector, residential cooking) except brick kilns remain shut down or less active late at night. This also explains why nighttime values of CO drop less in the winter and pre-monsoon than in other seasons. Furthermore, the prominent morning peaks of CO in pre-monsoon and winter compared to other seasons results from nighttime accumulation, additional fresh emissions in the morning and recirculation of the pollutants due to downslope katabatic winds (Pandey and Prinn, 2009; Panday et al., 2009). Pandey and Prinn (2009) observed nighttime accumulation and gradual decay during the winter (January 2005). The measurement site in Pandey and Prinn (2009) was near the urban core of the Kathmandu Valley and had significant influence from the vehicular sources all over the season including the winter season. Measurement in Bode lies in close proximity to the brick kilns which operate 24 hours during the winter and pre-monsoon period. Calm southeasterly winds are observed during the nighttime and early morning (ca.22:00 – 8:00) in pre-monsoon and winter, which transport emissions from brick kiln to the site (Sarkar et al., 2016). Thus the gradual decay in CO was not observed in Bode.

The timing of the CO morning peak observed in this study matches with observations by Panday et al. (2009). They also found CO morning peak at 8:00 in October 2004 and at 9:00 in January 2005. The difference could be linked to the boundary layer stability. As the sun rises later in
winter, the boundary layer stays stable for a longer time in winter keeping mixing ratios higher in morning hours than in other seasons with an earlier sunrise.

The morning peaks of CO\textsubscript{2} and CH\textsubscript{4} mixing ratios occurred around 6:00-7:00 local time in the pre-monsoon, monsoon, and post monsoon season, whereas in winter their peaks are delayed by 1-2 hours in the morning: CH\textsubscript{4} at 8:00 and CO\textsubscript{2} at 9:00. The CO showed that its morning peak was delayed compared to CO\textsubscript{2} and CH\textsubscript{4} morning peaks by 1-2 hour in pre-monsoon, monsoon and post-monsoon (at 8:00) and in winter (at 9:00). The occurrence of morning peaks in CO\textsubscript{2} and CH\textsubscript{4} 1-2 hours earlier than CO is interesting. This could be due to the long lifetimes and relatively smaller local sources of CH\textsubscript{4} and CO\textsubscript{2}, as CO is mainly influenced by emissions from vehicles during rush hour, as well as from biomass and trash burning in the morning hours. Also, CO increases irrespective of change in mixing layer (collapsing or rising, Figure 7) but CO\textsubscript{2} and CH\textsubscript{4} start decreasing only after the mixing layer height starts to rise. Recently, Chandra et al. (2016) also reported that the CO\textsubscript{2} morning peak occurred earlier than CO in observations in Ahmedabad City India. This was attributed to CO\textsubscript{2} uptake by photosynthetic activities after sunrise but CO kept increasing due to emissions from the rush hour activities.

The highest daytime minimum of CO\textsubscript{2} was observed in the pre-monsoon season followed by winter (Figure 6b), which may indicate the influence of regional emissions that increased the baseline background concentrations as well. The higher daytime minimum of CO\textsubscript{2} mixing ratios in the pre-monsoon season than in other seasons, especially winter, is interesting. The local emission sources are similar in pre-monsoon and winter and the boundary layer is higher (in the afternoon) during the pre-monsoon (~1200 meters) than in winter (~900 meters) (Mues et al., 2017). Occurs from 12:00 to 17:00 LST. The highest minimum CO\textsubscript{2} was found in pre-monsoon (Figure 6b). Although the local emission sources are similar in pre-monsoon and winter, the biospheric activity in the region is reported to be higher in the pre-monsoon (due to high temperature and solar radiation) than winter (Rodda et al., 2016). Among various possible causes, transport of CO\textsubscript{2} rich air from outside the Kathmandu Valley has been hypothesized as a main contributing factor, due to regional vegetation fire combined with westerly mesoscale to synoptic transport, the higher minimum daytime CO\textsubscript{2} mixing ratios in pre-monsoon season than other seasons, suggest the influence of regional emissions in the Kathmandu Valley, which has been reported in
previous study by Putero et al. (2015). In monsoon and post-monsoon seasons, the minimum CO₂ mixing ratios in the afternoon drops down to 390 ppm, which were close to the values observed at the regional background sites such as Mauna Loa and Waliguan.

3.4 Seasonal interrelation of CO₂, CH₄ and CO

The Pearson’s correlation coefficient (r) between CO₂ and CO was strong in winter (0.87), followed by monsoon (0.64), pre-monsoon (0.52) and post-monsoon (0.32). The higher coefficient in winter indicates that common or similar sources for CO₂ and CO and moderate values in pre-monsoon and monsoon indicates the likelihood of different sources. To avoid the influence of strong diurnal variations observed in the valley, daily averages, instead of hourly, were used to calculate the correlation coefficients. The correlation coefficients between daily CH₄ and CO₂ for four seasons are as follows: winter (0.80), post-monsoon (0.74), pre-monsoon (0.70) and monsoon (0.22). A semi-urban measurement study in India also found a strong positive correlation between CO₂ and CH₄ in the pre-monsoon (0.80), monsoon (0.61), post-monsoon (0.72) and winter (0.8) (Sreenivas et al., 2016). It should be noted here that Sreenivas et al., (2006) used hourly average CO₂ and CH₄ mixing ratios. The weak monsoon correlation at Bode, which is in contrast to Sreenivas et al. (2016), may point to the influence of dominant CH₄ emission from paddy field during the monsoon season (Goroshi et al., 2011). Daily CH₄ and CO was also weakly correlated in monsoon (0.34) and post-monsoon (0.45). Similar to CH₄ and CO₂, the correlation between CH₄ and CO were moderate to strong in pre-monsoon (0.76) and winter (0.75).

Overall, the positive and high correlations between CH₄ and CO mixing ratios and between CH₄ and CO₂ in the pre-monsoon and winter indicate common sources or source regions, most likely combustion related sources such as vehicular emission, brick kilns, agriculture fire etc., or the same source regions (i.e. their transport due to regional atmospheric transport mechanisms). Weak correlation, between CH₄-CO₂ and between CH₄-CO, during monsoon season indicates sources other than combustion-related may be active, such as agriculture as a key CH₄ source (Goroshi et al., 2013)
3.5 Influence of regional emission and transport

Regional sources and transport can influence the level of air pollution in the Kathmandu Valley mainly originating from regions west of the Kathmandu Valley (Putero et al., 2015). Wind from the north, which is less frequent than southerly and westerly winds, often brings cleaner air mass (also low in CH$_4$, Figure 4) and hence helps dilute or flush out the valley’s polluted air. Household combustion of biofuel, used mainly in the southern plains of Nepal and the IGP region, is an important contributor to the regional pollution in the higher mountainous areas (Panday and Prinn, 2009; Putero et al., 2014). Recently, Putero et al. (2015) attributed the afternoon high BC and O$_3$ concentrations at Paknajol in the Kathmandu Valley during pre-monsoon season to regional vegetation fire episodes and linked to the regional transport by westerly circulation. Our study also observed a number of episodes with high CO$_2$, CH$_4$ and CO mixing ratios at Bode during most of the days in March, April and May. During the entire sampling period of a year, there were 42 days with CO$_2$ mixing ratio $\geq$ 430 ppm, of which 29 days (or 69%) were during the pre-monsoon (25 days or 59% in March and April alone) and 10 days (23%) in winter. However, atmospheric chemistry transport models are required to confirm and differentiate contributions of local sources and regional sources influencing the Kathmandu Valley, which is beyond the scope of this study.

3.6.3.5 CO and CO$_2$ ratio: Potential emission sources

The ratio of the ambient mixing ratios of CO and CO$_2$ was used as an indicator to help discriminate emission sources in the Kathmandu Valley. The ratio was calculated from the excess (dCO and dCO$_2$) relative to the background values of ambient CO and CO$_2$ mixing ratios. The excess value was estimated by subtracting the base value which was calculated as the fifth percentile of the hourly data for a day (Chandra et al., 2016).

Average emission ratios from the literature are shown in Table 5, and average ratios of dCO/dCO$_2$ are shown in Table 6, disaggregated into morning hours, evening hours, and seasonal values. It must be stated that due to the large variance in the calculated ratio from this study.
(Table 6) as well as the likely variation in the estimated ratio presented in Table 5, the
interpretation and conclusion about sources should be cautiously drawn and will be indicative.
Higher ratios were found in pre-monsoon (12.4) and winter (15.1) season compared to post-
monsoon (8.3) and monsoon (7.5). These seasonal differences in the dCO/dCO\textsubscript{2} ratio are
depicted in Figure 8, which shows a clear relationship with the wind direction and associated
emissions, with the highest values especially for stronger westerly winds. Compared to the other
three seasons, the ratio in winter was also relatively high for air masses from the east, likely due
to emissions from brick kilns combined with accumulation during more stagnant meteorological
conditions (supplementary information Figure S2, S3). In other seasons, emission emanating
from the north and east of Bode were characterized by a dCO/dCO\textsubscript{2} ratio below 15. Air masses
from the west and south generally have a ratio from 20 to 50 in all but post-monsoon season,
where the ratio sometimes exceeds 50. A ratio of 50 or over is normally due to very inefficient
combustion sources (Westerdahl et al., 2009; Stockwell et al., 2016), such as agro-residue
burning, which is common during the post-monsoon season in the Kathmandu Valley.

For interpretability of emission ratio with sources, the ratio was classified into three categories:
(i) 0 – 15, (ii) 15 – 45, and (iii) greater than 45. This classification was based on the observed
distribution of emission ratio during the study period (Figure 8) and a compilation of observed
emission ratios typical for different sources from Nepal and India (see Table 5). An emission
ratio below 15 is likely to indicate residential cooking and diesel vehicles, and captive power
generation with diesel-powered generator sets (Smith et al., 2000; ARAI, 2008; World Bank,
2014). The emission from brick kilns (FCBTK and Clamp kilns, both common in the Kathmandu
Valley), and inefficient, older (built before 2000) gasoline cars fall in between 15 - 45 (Weyant
et al., 2014, Stockwell et al., 2016; ARAI, 2008). Four-stroke motorbikes and biomass burning
activities (mixed garbage, crop-residue and biomass) are one of the least efficient combustion
sources, with emission ratios higher than 45 (Westerdahl et al., 2009; Stockwell et al., 2016;
ARAI, 2008).

Based on the classification and Figure 8, the emissions from sources to the north and east of the
site are dominated by residential cooking and/or diesel combustion. Emissions from the south
and west of Bode are mainly contributed by sources such as brick kilns and inefficient gasoline
vehicles. Very high ratios, indicative of agro residue open burning, generally only show up during the post-monsoon period, when such activities take place, especially in areas southwest of the site. The relatively enhanced ratio (20-30) observed in winds from north and east of the site during winter is mostly likely due to brick kilns that use mixed coal biomass fuel, whereas the Figure 8 indicates the dominant signature of residential cooking, diesel and old gasoline cars during the pre-monsoon, monsoon and post-monsoon seasons. Although ratio of CO/CO2 is a weak indicator of sources and the mean ratio has large variance (See Table 6), the conclusions drawn, from using Figure 8 and the above mentioned classification, are not conclusive. The estimated CO/CO2 ratio tentatively indicates that the local plume impacting the measurement site (Bode) from the north and east could be residential and/or diesel combustion. The estimated CO/CO2 ratio of the local plume from the south and west generally falls in the 15-45 range which could indicate emissions from brick kilns and inefficient gasoline vehicles. Very high ratios were also estimated from the south west during the post-monsoon season. Among other possible sources, this may indicate agro-residue open burning.

The emission inventory for CO identifies (aggregate for a year) residential, and gasoline related emission from transport sector (Sadavarte et al., 2017, in preparation). The inventory is not yet temporally resolved, so no conclusion can be drawn about the sources with respect to different seasons. From the 1km x1km emission inventory of the Kathmandu Valley for 2011, the estimated sectoral source apportionment of CO is residential (37%), transport sector (40%) and industrial (20%). The largest fraction from the residential sector is cooking (24 %) whereas the majority of transport sector related CO in the Kathmandu Valley is from gasoline vehicles.

The dCO/dCO2 ratio also changes markedly between the morning peak hours (7:00-9:00, except in winter season when the peak occurs during 8:00-9:00) and evening peak hours (19:00-21:00 pm) (Table 6). Morning and evening values were lowest (2.2, 8.0) during the monsoon and highest (11.2, 21.6) in the winter season, which points to the different emission characteristics in these two seasons. This feature is similar to Ahmedabad, India, another urban site in south Asia, where the morning/evening values were lowest (0.9/19.5) in monsoon and highest in winter (14.3/47.2) (Chandra et al., 2016). In the morning period, the ratio generally falls within a narrower range, from less than 1 to about 25, which indicates a few dominant sources, such as
cooking, diesel vehicles, and diesel gen-sets (see Figure 9). In the evening period, the range of the ratio is much wider, from less than 1 to more than 100, especially in winter. This is partly due to the shallower boundary layer in winter, giving local CO emissions a chance to build up more rapidly compared to the longer-lived and well-mixed CO2, and also indicating the prevalence of additional sources such as brick kilns and agro-residue burning.

3.6.7 Comparison of CH4 and CO2 at semi-urban site (Bode) and rural site (Chanban)

Figure 10 shows time series of hourly average mixing ratios of CH4, CO2, CO and water vapor observed simultaneously at Bode and Chanban for the period of 15th July to 3rd October 2015. The hourly meteorological parameters observed at Chanban are shown in supplementary Figure S4. The hourly temperature ranges from 14 to 28.5 °C during the observation period. The site experienced calm winds during the night and moderate southeasterly winds with hourly maximum speed of up to 7.5 m s⁻¹ during the observation period. The CH4 mixing ratios at Chanban varied from 1.880 ppm to 2.384 ppm, and generally increased from the last week of July until early September, peaking around 11th September and then falling off towards the end of the month. CO followed a generally similar pattern, with daily average values ranging from 0.10 ppm to 0.28 ppm. The hourly CO2 mixing ratios ranged from 375 to 453 ppm, with day to day variations, but there were no clear pattern as observed in trend like CH4 and CO mixing ratios.

The CH4, CO2, and CO mixing ratios were higher in Bode than in Chanban (Figure 10, Table 4), with Chanban approximately representing the baseline of the lower envelope of the Bode levels. The mean CO2, CH4 and CO mixing ratios over the entire sampling period of nearly three months at Bode are 3.8%, 12.1%, and 64% higher, respectively, than at Chanban. The difference in the CO2 mixing ratio could be due to the large uptake of CO2 in the forested area at Chanban and surrounding regions compared to Bode, where the local anthropogenic emissions rate is higher and less vegetation for photosynthesis. The coincidence between the base values of CO and CH4 mixing ratios at Bode and the levels observed at Chanban implies that Chanban CO and CH4 mixing ratios are indicative of the regional background levels. A similar increase in CO and CH4 mixing ratios at Chanban from July to September was also observed at Bode, which may
imply that the regional/background levels in the broader Himalayan foothill region also influences the baseline of the daily variability of the pollutants in the Kathmandu Valley, consistent with Panday and Prinn (2009).

Figure 11 shows the comparison of average diurnal cycles of CO₂, CH₄, CO and water vapor mixing ratios observed at Bode and Chanban. The diurnal pattern of CO₂ mixing ratios at both sites is similar, but more pronounced at Bode, with a morning peak around 6:00-7:00, a daytime minimum, and a gradual increase in the evening until the next morning peak. A prominent morning peak at Bode during the monsoon season indicates the influence of local emission sources. The daytime CO₂ mixing ratios are also higher at Bode than at Chanban because of local emissions less uptake of CO₂ for photosynthesis in the valley in comparison to the forested area around Chanban. Like the diurnal pattern of CO₂ depends on the evolution of the mixing layer at Bode, as discussed earlier, it is expected that the mixing layer evolution similarly influences the diurnal CO₂ mixing ratios at Chanban. CO, on the other hand, shows very different diurnal patterns at Bode and Chanban. Sharp morning and evening peaks of CO are seen at Bode, indicating the strong local polluting sources, especially cooking and traffic in the morning and evening peak hours. Chanban, in contrast, only has a subtle morning peak and no evening peak. After the morning peak, CO sharply decreases at Bode but not at Chanban. The growth of the boundary layer after sunrise and entrainment of air from the free troposphere, with lower CO mixing ratios, causes CO to decrease sharply during the day at Bode. At Chanban, on the other hand, since the mixing ratios are already more representative of the local and regional background levels which will also be prevalent in the lower free troposphere, CO does not decrease notably during the daytime growth of the boundary layer as observed at Bode.

Similarly, while there is very little diurnal variation in the CH₄ mixing ratios at Chanban, there is a strong diurnal cycle of CH₄ at Bode, similar to CO₂ there. At Chanban, the CH₄ mixing ratio only shows a weak minimum at around 11 am, a slow increase during the day until a its peak around 22:00, followed by a slow decrease during the night and a more rapid decrease through the morning. The cause of this diurnal pattern at Chanban is presently unclear, but it is clear that the levels could be generally representative of the regional background throughout the day and show only limited influences of local emissions.
4. Conclusions

A cavity ring down spectrometer (Picarro G2401, USA) was used to measure ambient CO₂, CH₄, CO, and water vapor mixing ratios at a semi-urban site (Bode) in the Kathmandu Valley for a year. This was the first 12-months of continuous measurements of these four species in the Kathmandu Valley in the foothills of the central Himalaya. Simultaneous measurement was carried out at a rural site (Chanban) for approximately 3 months to evaluate urban-rural differences.

The measurement also provided an opportunity to establish diurnal and seasonal variation of these species in one of the biggest metropolitan cities in the foothills of Himalayas. Annual average of the mixing ratio of CH₄ and CO₂ in Bode revealed that they were higher than the concentrations mixing ratios at the background sites such as the Mauna Loa, USA and Mt. Waliguan, China, as well as higher than urban/semi-urban sites in nearby regions such as Ahmedabad and Shadnagar in India, and Nanjing in China. These comparisons highlight potential sources of CH₄ and CO₂ in the Kathmandu Valley, such as brick kilns in the valley.

Polluted air masses were transported to the site mainly by two major local wind circulation patterns, East-South/North East and West-Southwest throughout the observation period. Strong seasonality was observed with CO compared to CO₂ and CH₄. Winter and pre-monsoon high CO are linked to emission sources active in these seasons only and are from east-southeast and west-southwest. Emission from the east-southeast are most likely related to brick kilns (winter and pre-monsoon), which are in close proximity to Bode. Major city-centers are located in the west-southwest of Bode (vehicular emission) which impact the site all-round the year, although higher during winter season. Winter high was also observed with CO₂ and CH₄, which are mostly local influence of brick kilns, trash burning and emission from city-center. Nighttime and early morning accumulation of pollutants in winter due to a shallow stable mixing height (ca. 200 m) also contribute to elevated levels than other seasons. Regional transport into the Kathmandu Valley could be related to CO₂ peak during pre-monsoon. The highest CH₄ during the post-monsoon could be associated with agricultural activity northeast of Bode. Diurnal variation across all seasons indicates the influence of rush-hour emissions related to vehicles and
residential emissions. The evolution of the mixing layer height (200-1200 m) was a major factor which controls the morning-evening peak, afternoon low and night-early morning accumulation or decay. Thus the geographical setting of the Kathmandu Valley and its associated meteorology play a key role in the dispersion and ventilation of pollutants in the Kathmandu Valley. The ratio of CO/CO₂ across different season and wind direction showed that emissions from inefficient gasoline vehicles, brick kilns, residential cooking and diesel combustion are likely to impact Bode.

The differences in mean values for urban-rural measurements at Bode and Chanban is highest for CO (64 %) compared to CO₂ (3.8%) and CH₄ (12%). Low values of CH₄ and CO₂ mixing ratios at the Chanban site could represent a regional background mixing ratios.

This study has provided valuable information on key greenhouse gases and air pollutants in the Kathmandu Valley and the surrounding regions. These observations can be useful as ground-truthing for evaluation of satellite measurements, as well as climate and regional air quality models. The overall analysis presented in the paper will contribute along with other recent measurement and analysis to can providing e a sound scientific basis for reducing emissions of greenhouse gases and air pollutants in the Kathmandu Valley.

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References


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<th>Site</th>
<th>Instrument</th>
<th>Species</th>
<th>sampling interval</th>
<th>Measurement period</th>
<th>inlet/sensor height above ground (m)</th>
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<td>i. Cavity ring down spectrometer (Picarro G2401, USA)</td>
<td>CO₂, CH₄, CO, water vapor</td>
<td>5 sec</td>
<td>06 Mar 2013 - 05 Mar 2014</td>
<td>20</td>
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<td></td>
<td>ii. CO monitor (Horriba AP370, USA)</td>
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<td>5 min</td>
<td>06 Mar 2013 – 07 June 2013</td>
<td>20</td>
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<td></td>
<td>iii. Ceilometer (Vaisala CL31, Finland)</td>
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<td>15-52 min</td>
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<td></td>
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<td>1 min</td>
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<td>b. CS300 Pyranometer</td>
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<td>c. RM Young 05103-5</td>
<td>WD, WS</td>
<td></td>
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<td>v. Airport AWS (Environdata, Australia)</td>
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<td>a. TA10</td>
<td>T</td>
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<td>18 Jun 2013 – 13 Jan 2013</td>
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<td>b. RG series</td>
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<td>5 sec</td>
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<td></td>
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Table 2. Summary of monthly average CH$_4$ and CO$_2$ mixing ratios observed at Bode, a semi-urban site in the Kathmandu Valley during March 2013 to Feb 2014 [mean, standard deviation (SD), median, minimum (Min.), maximum (Max.) and number of data points of hourly average values]

<table>
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<tr>
<th>Month</th>
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<th></th>
<th></th>
<th></th>
<th>CO$_2$ (ppm)</th>
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<th></th>
<th></th>
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<td></td>
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<td>Min.</td>
<td>Max.</td>
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<td>SD</td>
<td>Median</td>
<td>Min.</td>
<td>Max.</td>
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<td>418.3</td>
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<td>510.8</td>
<td>596</td>
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<td>2.094</td>
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<td>713</td>
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<td>390.5</td>
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<td>30.2</td>
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Table 3. Summary of CH$_4$ and CO$_2$ mixing ratios at Bode across four seasons during March 2013 to Feb 2014 [seasonal mean, one standard deviation (SD), median, minimum (Min.) and maximum (Max.).]

<table>
<thead>
<tr>
<th>Season</th>
<th>CH$_4$ (ppm)</th>
<th></th>
<th></th>
<th></th>
<th>CO$_2$ (ppm)</th>
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<tbody>
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<td></td>
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Table 4. Comparison of monthly average CH$_4$ and CO$_2$ mixing ratios at a semi-urban and a rural site in Nepal (this study) with other urban and background sites in the region and elsewhere.

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<tr>
<th>Site</th>
<th>Setting</th>
<th>Bode, Nepal (Urban)</th>
<th>Chanban, Nepal (Rural)</th>
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<tr>
<td>Species</td>
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<td>Annual CO₂ (ppm)</td>
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<tr>
<td>Mauna Loa</td>
<td></td>
<td></td>
<td>396.8</td>
<td>1.832</td>
<td></td>
</tr>
<tr>
<td>Waliguan</td>
<td></td>
<td></td>
<td>397.7</td>
<td>1.880</td>
<td></td>
</tr>
<tr>
<td><strong>Nanjing (2011)</strong></td>
<td></td>
<td>406.5</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Shadnagar (2014)</td>
<td></td>
<td>394.0</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ahemadabad (2013-2015)</td>
<td>413.0</td>
<td>1.920</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>


Table 5. Emission ratio of CO/CO₂ (ppb ppm⁻¹) derived from emission factors (gram of gas emitted from per kilogram of fuel burned, except transport sector which is derived from gram of gases emitted per kilometer distance travelled)

<table>
<thead>
<tr>
<th>Sectors</th>
<th>Details</th>
<th>CO/CO₂</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Residential/Commercial</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>i. LPG</td>
<td></td>
<td>4.8</td>
<td>Smith et al. (2000)</td>
</tr>
<tr>
<td>ii. Kerosene</td>
<td></td>
<td>13.4</td>
<td>Smith et al. (2000)</td>
</tr>
<tr>
<td>iii. Biomass</td>
<td></td>
<td>52.9 - 98.5</td>
<td>*</td>
</tr>
<tr>
<td>iv. Diesel power generators</td>
<td>&lt; 15 year old</td>
<td>5.8</td>
<td>The World Bank (2014)</td>
</tr>
<tr>
<td></td>
<td>&gt;15 year old</td>
<td>4.5</td>
<td></td>
</tr>
<tr>
<td>2. Transport</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>a. Diesel</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>i. HCV diesel bus</td>
<td>&gt;6000cc, 1996-2000</td>
<td>4.9</td>
<td></td>
</tr>
<tr>
<td></td>
<td>post 2000 and 2005</td>
<td>5.4</td>
<td></td>
</tr>
<tr>
<td>ii. HCV diesel truck</td>
<td>&gt;6000cc, post 2000</td>
<td>7.9</td>
<td></td>
</tr>
<tr>
<td>b. Petrol</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>i. 4 stroke motorcycle</td>
<td>&lt;100 cc, 1996-2000</td>
<td>68</td>
<td></td>
</tr>
<tr>
<td></td>
<td>100-200 cc, Post 2000</td>
<td>59.6</td>
<td></td>
</tr>
<tr>
<td></td>
<td>&lt;1000 cc, 1996-2000</td>
<td>42.4</td>
<td></td>
</tr>
<tr>
<td>ii. Passenger cars</td>
<td>2000</td>
<td>10.3</td>
<td></td>
</tr>
<tr>
<td>iii. Passenger cars</td>
<td>&lt;1000 cc, Post 2000</td>
<td></td>
<td></td>
</tr>
<tr>
<td>3. Brick industries</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>i. BTK fixed kiln</td>
<td></td>
<td>17.2</td>
<td>Weyant et al. (2014)</td>
</tr>
<tr>
<td>ii. Clamp brick kiln</td>
<td></td>
<td>33.7</td>
<td>Stockwell et al. (2016)</td>
</tr>
<tr>
<td>iii. Zigzag brick kiln</td>
<td></td>
<td>3.9</td>
<td>Stockwell et al. (2016)</td>
</tr>
<tr>
<td>4. Open burning</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>i. Mixed garbage</td>
<td></td>
<td>46.9</td>
<td>Stockwell et al. (2016)</td>
</tr>
<tr>
<td>ii. Crop-residue</td>
<td></td>
<td>51.6</td>
<td>Stockwell et al. (2016)</td>
</tr>
</tbody>
</table>

* Westerdahl et al. (2009)
** http://www.cpcb.nic.in/Emission_Factors_Vehicles.pdf
Table 6. Seasonal average (SD) of the ratio of dCO to dCO₂, their Geometric mean (GeoSD) over a period of 3 hours during (a) morning peak and (b) evening peak and (c) seasonal (all hours) of the ambient mixing ratios of CO and CO₂. And their lower and upper bound (LB and UB).

<table>
<thead>
<tr>
<th>Period</th>
<th>Season</th>
<th>Mean (SD)</th>
<th>Median</th>
<th>N</th>
<th>Geomean (GeoSD)</th>
<th>LB</th>
<th>UB</th>
</tr>
</thead>
<tbody>
<tr>
<td>a. Morning hours</td>
<td>Pre-monsoon</td>
<td>7.6 (3.1)</td>
<td>7.8</td>
<td>249</td>
<td>11.3 (1.5)</td>
<td>5.2</td>
<td>24.8</td>
</tr>
<tr>
<td></td>
<td>Monsoon</td>
<td>2.2 (1.6)</td>
<td>1.9</td>
<td>324</td>
<td>9.9 (1.9)</td>
<td>2.7</td>
<td>36.3</td>
</tr>
<tr>
<td></td>
<td>Post-monsoon</td>
<td>3.1 (1.4)</td>
<td>2.8</td>
<td>183</td>
<td>11.1 (1.5)</td>
<td>4.7</td>
<td>26.3</td>
</tr>
<tr>
<td></td>
<td>Winter*</td>
<td>11.2 (4.4)</td>
<td>11</td>
<td>255</td>
<td>11.4 (1.5)</td>
<td>5.3</td>
<td>24.2</td>
</tr>
<tr>
<td>b. Evening hours</td>
<td>Pre-monsoon</td>
<td>15.1 (9.0)</td>
<td>12.7</td>
<td>248</td>
<td>10.5 (1.7)</td>
<td>3.5</td>
<td>31.6</td>
</tr>
<tr>
<td></td>
<td>Monsoon</td>
<td>8.0 (5.2)</td>
<td>6.3</td>
<td>323</td>
<td>10.2 (1.8)</td>
<td>3.1</td>
<td>33.5</td>
</tr>
<tr>
<td></td>
<td>Post-monsoon</td>
<td>11.5 (5.6)</td>
<td>10.6</td>
<td>182</td>
<td>11.0 (1.6)</td>
<td>4.4</td>
<td>27.6</td>
</tr>
<tr>
<td></td>
<td>Winter</td>
<td>21.6 (14.1)</td>
<td>18.2</td>
<td>254</td>
<td>10.2 (1.8)</td>
<td>3.1</td>
<td>33.6</td>
</tr>
<tr>
<td>c. Seasonal</td>
<td>Pre-monsoon</td>
<td>12.2 (13.3)</td>
<td>8.8</td>
<td>1740</td>
<td>8.2 (2.4)</td>
<td>1.4</td>
<td>48.4</td>
</tr>
<tr>
<td></td>
<td>Monsoon</td>
<td>7.5 (13.5)</td>
<td>2.9</td>
<td>2176</td>
<td>5.9 (3.3)</td>
<td>0.5</td>
<td>65.6</td>
</tr>
<tr>
<td></td>
<td>Post-monsoon</td>
<td>8.3 (12.4)</td>
<td>4.4</td>
<td>1289</td>
<td>6.8 (3.0)</td>
<td>0.8</td>
<td>59.2</td>
</tr>
<tr>
<td></td>
<td>Winter</td>
<td>15.1 (13.3)</td>
<td>12.5</td>
<td>1932</td>
<td>9.2 (2.1)</td>
<td>2.0</td>
<td>41.7</td>
</tr>
</tbody>
</table>

*The morning peak was one hour delayed in winter, thus the 8:00-10:00 period data was used in the analysis.
Figure 1. Location of measurement sites: (a) Kathmandu Valley (b) semi-urban measurement site at Bode in Kathmandu Valley, and a rural measurement site at Chanban in Makawanpur district Nepal, (c) general setting of Bode site. Colored grid and TIA represent population density and the Tribhuvan International Airport, respectively.
Figure 2. Time series of hourly average (a) mixing ratios of CH$_4$, CO$_2$, CO, and water vapor measured with a cavity ring down spectrometer (Picarro G2401) at Bode, and (b) temperature and rainfall monitored at the Tribhuvan International Airport (TIA), ~4 km to the west of Bode site in the Kathmandu Valley, Nepal. Temperature shown in pink color is observed at Bode site.
Figure 3. Monthly variations of the mixing ratios of hourly (a) CH₄, (b) CO₂, (c) CO, and (d) water vapor observed at a semi-urban site (Bode) in the Kathmandu Valley over a period of a year. The lower end and upper end of the whisker represents 10ᵗʰ and 90ᵗʰ percentile, respectively; the lower end and upper end of each box represents 25ᵗʰ and 75ᵗʰ percentile, respectively, and black horizontal line in the middle of each box is the median for each month while red dot represents mean for each month.
Figure 4. Relation between Pollution rose of the hourly CH₄ and CO₂ mixing ratios and wind direction observed at Bode in the Kathmandu Valley (a) CH₄ and (b) CO₂ and (c) CO from March 2013 to February 2014. The figure shows variations of pollutants CH₄, CO₂ and CO mixing ratios based on frequency of counts by wind direction (in %) as represented by circle. The color represents the different mixing ratios of the gaseous species. The units of CH₄, CO₂ and CO are in ppm.
Figure 5. Satellite detected fire counts in (a) Mar, (b) Apr, (c) May 2013 in the broader region surrounding Nepal and (d) total number of fire counts detected by MODIS instrument onboard the Aqua satellite during Jan 2013-Feb 2014. Source: https://firms.modaps.eosdis.nasa.gov/firemap/
Figure 6. Diurnal variations of hourly mixing ratios in different seasons (a) CH$_4$, (b) CO$_2$, (c) CO, and (d) water vapor observed at Bode (semi-urban site) in the Kathmandu Valley during March 2013-February 2014. Seasons are defined as Pre-monsoon: Mar-May, Monsoon: Jun-Sep, Post-monsoon: Oct-Nov, Winter: Dec-Feb. The x axis is in Nepal Standard Time (NST).
Figure 7. Diurnal variations of hourly mixing ratios of CH₄, CO₂, CO, and mixing layer height (MLH) at Bode (a semi-urban site in the Kathmandu Valley) in different seasons (a) pre-monsoon (Mar-May), (b) monsoon (Jun-Sep), (c) post-monsoon (Oct-Nov) and (d) winter (Dec-Feb) during March 2013- Feb 2014.
Figure 8. Seasonal polar plot of hourly dCO/dCO$_2$ ratio based upon wind direction and wind speed: (a) pre-monsoon, (b) monsoon, (c) post-monsoon and (d) winter seasons.
Figure 9. Seasonal frequency distribution of hourly dCO/dCO₂ ratio (a) morning hours (7:00-9:00) in all season except winter (8:00-10:00), (b) evening hours (19:00-21:00)
Figure 10. Comparison of hourly average mixing ratios of CH₄, CO₂, CO, and water vapor observed at Bode (a semi-urban site) in the Kathmandu Valley and at Chanban (a rural/background site) in Makawanpur district, ~ 20 km from Kathmandu, on other side of a tall ridge.
Figure 11. Diurnal variations of hourly average mixing ratios of (a) CH$_4$, (b) CO$_2$, (c) CO and (d) water vapor observed at Bode in the Kathmandu Valley and at Chanban in Makawanpur district during 15 July- 03 October 2015.